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LES ELEMENTS ET LEURS ISOTOPES DANS L'UNIVERS

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ISOTOPIC HETEROGENEITIES IN THE SOLAR SYSTEM

SPECIAL REPORT

G.J. WASSERBURG, D.A. PAPANASTASSIOU, and TYPHOON LEE
The Lunatic Asylum of the Charles Arms Laboratory
Division of Geological and Planetary Sciences *
California Institute of Technoloy
Pasadena, California 91125, U.S.A.

ABSTRACT

The nature of isotopic anomalies in solar system material is discussed with emphasis on correlated anomalies in refractory elements. Evidence for the existence of short-lived radioactive nuclides in the early solar system is given and is used to estimate the time scale for the last injection of freshly synthesized nuclear material. It is shown that the early solar nebula was incompletely mixed and contained debris which was injected from a stellar source at most a few million years prior to the formation of the solar system. It appears that the average solar system material is made up of ambient interstellar material which was deficient in certain nuclear species to which was added a small fraction of freshly synthesized material. It is this mixture which now makes up the bulk solar system. At the present time there may still remain both substantial and subtle isotopic differences between the sun, the terrestrial planets, and volatile-rich planetary bodies including comets. The isotopic variations which are observed reflect slightly different proportions of nuclei from different stellar sources which were locally well mixed and homogenized prior to or during the formation of the early solar nebula condensates. The process of local homogenization apparently destroyed most pre-solar dust grains but preserved the distinctive average isotopic character of the local regions. Substantial isotopic differences existed between the cool gas and some condensed matter. This is manifest in major chemical and some associated isotopic alteration of the early condensates.

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INTRODUCTION

It has now been established that the isotopic composition of many elements in some meteoritic materials is distinctly different from that in terrestrial samples and that very short-lived radioactive nuclides were present in the early solar system. These discoveries indicate that we now have the possibility of identifying and studying the specific astrophysical and cosmochemical processes which immediately preceded the formation of the sun and the solar system. The solar system was formed from a mixture of gas and dust *about* 4.6 AE ago. In general, the sequence of formation of the planets and the sun is not known. At 4.54 AE ago some small planetary objects melted and differentiated. At about 4.45 AE ago the earth and the moon were involved in a major chemical differentiation process. The determination of these ages is fundamentally dependent on the existence of chemical fractionation between the radioactive parent and stable daughter elements. The ages are thus the times of chemical fractionation. This difference of 0.1 AE may represent differentiation processes within the parent planets or possibly the time required to aggregate the larger terrestrial planets from the smaller debris in the inner solar system. To correlate this aggregation process with the 0.1 AE time difference requires that substantial fractionation of volatile from involatile elements take place during the final accretion events. Intervals of 0.1 AE for accretion of the planets have been argued from theoretical considerations (*cf.* GOLDREICH and WARD, 1973; SAFRONOV, 1969). The postaccretional history of the terrestrial planets is marked by planetary differentiation processes and a decreasing intensity of bombardment by planetary debris. The late (postaccretional) bombardment history was indexed by an intense and well-defined event on the moon at 3.9 AE which has been associated with the major cratering of the whole inner solar system (Mars, Earth-Moon, Venus, and Mercury; TERA, PAPANASTASSIOU and WASSERBURG, 1974). The subsequent planetary evolution has been governed by internal differentiation supported by heat sources. This planetary differentiation was most intense on the earth but has occurred to varying degrees on all of the terrestrial planets. The particular history of the earth for the past 3.9 AE is still being unraveled by use of old and new techniques and theories. However, the very early history of the earth is not known. The post-accretional histories of the other planets are just now being addressed in the framework of a broad-scale exploration of the solar system which involves the use of spacecraft. In searching for the clues about the very early history of the solar system just before 4.555 AE the keys have come from studies of meteorites. Meteorites which come from small planetary objects (less than ~ 100 km) provide relics of very ancient solar system processes as they are less subject to the more complex internal evolution which continues to occur on larger planetary bodies.

The meteorites comprise a highly diverse group of objects that includes the results of melting and chemical-mechanical segregation on small

planets and those materials (the chondrites) which appear to be aggregates of more primitive debris. The relative abundances of the chemical elements in the chondrites have been used as a basis for estimating the solar abundances (relative to Si). In particular, the type I carbonaceous chondrites which are relatively rich in volatile elements have provided one of the precise measures of the relative abundances of the elements. These abundances compare very closely with those which can be reliably measured from spectroscopic studies of the solar atmosphere. Knowledge of the isotopic abundances and of the chemical abundances gives the abundances of the nuclear species in the « sun » and has provided the fundamental data base for all theories of the formation of the elements. All the precise isotopic abundance data have come from measurements of terrestrial samples, meteorites and, subsequent to the Apollo missions, lunar samples. Good precision data on isotopic abundances for a few elements have been obtained for the sun, and these are in reasonable agreement with the observations on terrestrial materials.

Some time ago, it was recognized that the observed abundances of the nuclear species in the sun are primarily the result of nuclear reactions in a variety of environments inside different stars. The concept that the solar abundances were the average of a variety of nuclear processes in numerous stellar sources was a major advance in understanding the origin of the elements. Some of the abundant nuclear species such as ^{4}He, ^{12}C, ^{16}O, ^{20}Ne, ^{28}Si, and ^{56}Fe are the direct product of hydrostatic nuclear burning and are related to the major energy sources governing stellar evolution. These species are produced under conditions which are reasonably well understood and are associated with well-defined steliar sites. Other nuclear species are considered to be produced by modification of the production pattern of hydrostatic burning during transient explosive phenomena upon the death of massive stars. These processes appear to be important but have not been clearly understood in the context of realistic stellar environments. Often the abundance of minor nuclear species are not clearly constrained by our knowledge of stellar evolution and may be affected by several distinct burning events. The production of such nuclides is at present not well known and cannot be associated with particular sites within stars. The mechanisms which contribute to the formation of a particular nuclear species are thus complex and depend upon a thorough knowledge of the nuclear reaction rates, time scales, and an understanding of the different possible stellar sites in which the appropriate reactions may go on. Furthermore, the details of the ejection of processed matter back into the interstellar medium and the recycling of these materials in new generations of stars are not well understood.

One of the most important results to come out of studies of the abundances of the nuclear species in the solar system, which are widespread through the population I stars, is the classification of the nuclei into *s*, *r*, and *p* types. This discussion follows the pioneer work of BURBIDGE, BURBIDGE, FOWLER and HOYLE (1957) and CAMERON (1957). The

s-type nuclei may be produced by a continuous chain of neutron captures starting with around $Z = 26$ (Fe) and extending up to ^{209}Bi. These nuclei are connected by neutron capture followed by β decay. The rates of neutron capture are assumed to be sufficiently slow so that β decays with a lifetime of ~ 10 years may take place without a significant opportunity for the nucleus capturing another neutron. The *r*-type nuclei (see Figure 1) are unshielded, neutron-rich nuclei and would be the result of the β decay of very neutron-rich unstable precursor nuclei. The *r* nuclei also include U and Th and all of the elements heavier than ^{209}Bi. In fact, it is the existence of all of these long-lived heavier elements, that are isolated from other stable nuclear species in the chart of the nuclides, which most clearly separates the *r* and *s* nuclei and defines the need for such a classification. Extensive theoretical discussions on these processes have been given in the literature.

Some nuclear species may be considered as both *s* and *r* types (see Figure 1). The neutron-rich precursors of the *r* type nuclei are required to

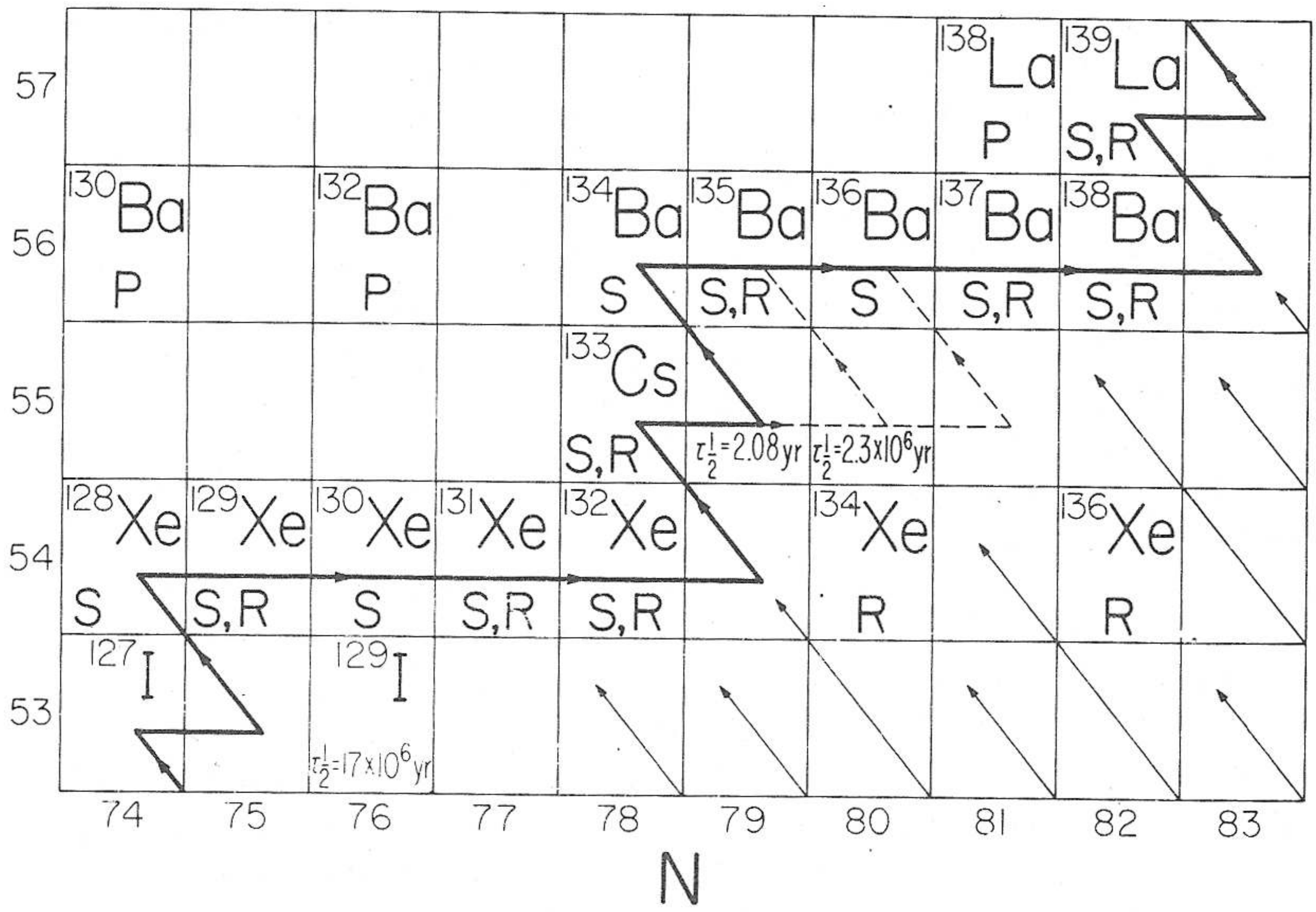


Fig. 1. Chart of the nuclides showing the *s*-process path (heavy line) in the vicinity of Xe and Ba. Isotopes on the path are reached by neutron captures on a timescale slow compared to the half-lives for β decay along the path. Isotopes on the proton rich side of the path (*e.g.*, ^{130}Ba, ^{132}Ba) are bypassed by the *s*-process and require a separate, *p*-process (*p*, γ or γ, *n* reactions). Isotopes on the neutron rich side of the path are also bypassed by the *s*-process and are produced by the successive β decays of their more neutron-rich isobars produced by neutron captures on a timescale which is rapid compared to β decays (*r*-process). Note isotopes shielded from the *r*-process and produced only by the *s*-process (*e.g.*, $^{134, 136}$Ba).

form in a neutron-rich environment at high temperatures and densities and may only exist for a short time (~ 1s). The astrophysical sites in which the *r* type nuclei may be produced have never been clear but the strongest association has been with supernova events. It is quite possible that the «classical *r*» process with $(n, \gamma) \rightleftharpoons (\gamma, n)$ quasi-equilibrium is not the true means of producing «*r*» process nuclei (TRURAN, COWAN and CAMERON, 1978). The *p* type nuclei are neutron-poor isotopes which cannot be reached by the *s* chain and are typically rather rare. These have sometimes been considered as resulting from proton bombardment of pre-existing *s* and *r* nuclei or by (γ, n) reactions, but neither the nuclear processes nor the astrophysical sites associated with these processes have been clear (*cf.* WOOSLEY and HOWARD, 1978).

There are a few other nuclei which cannot be classified in this manner, but none for $Z \geq 26$. The *s*, *r*, *p* classification has proven to be a basic one which is fundamental to any discussion of the formation of the elements and hence nuclei are called *s*, *r*, *p* «process». However, the *s*, *r* and *p* processes referred to are the conceptual ones outlined above and are not as yet related to actual nuclear-astrophysical processes and sites. The solar system abundances are believed to be averages of the production of the nuclear species through several different stellar sources and many cycles of processing and injection into the interstellar medium and reprocessing.

In order to understand the processes involved in generating the elements, it is necessary to utilize both astronomical observations which directly or indirectly indicate nucleosynthetic activity and to pursue, through theoretical considerations, the hints contained in the abundances of the nuclear species and in models of stellar evolution. Indeed, if it were possible to obtain samples of matter from distinctive stellar sources, then our understanding of the mechanisms of element generation could be greatly advanced. For over two decades it has been clear that different stellar components are present in the material which makes up the solar system. The possibility that some pre-solar system star-dust has been preserved until today has long been a matter of inquiry. The preservation of pre- or proto-solar gas and dust requires that the mixing processes in the interstellar medium and in the formation of the solar system be incomplete. It further requires that the physical and chemical differentiation processes within the solar system over the past ~ 4.6 AE not erase the isotopic and chemical signatures of the precursor sources. Until recently, the dominant working hypothesis in the study of nucleosynthesis and of the formation of the solar system was that the solar system was isotopically homogeneous but chemically heterogeneous as a result of chemical fractionation. There was no evidence for the survival of identifiable components of pre-solar matter. From the discovery of ^{129}I present in meteorites it was concluded that the last injection of freshly-synthesized nuclear material took place $\sim 1.6 \times 10^8$ y before the formation of the solar system. This viewpoint has now been changed as a result of a series of discoveries over the past few years.

THE MAGIC OF METEORITES

Meteorites are derived from relatively small planetary objects ($\lesssim$ 100 km) which are thought to reside in the asteroidal belt and possibly some in the Oort cometary cloud. These bodies are subject to physical disruption and gravitational perturbations which cause them to collide with the earth and other planets. Subsequent to disruption and breakup, they are known to survive typically for only a few million years and some as long as $\sim 5 \times 10^8$ y (BEGEMANN, GEISS and HESS, 1957; VOSHAGE, 1978). The nature of the meteoritic debris which falls on the earth shows it to be comprised of two distinctive classes. The first are aggregates of varied materials, often full of ovoidal pellets (chondrules) which appear to be droplets of some kind, imbedded in a fine-grained matrix. These meteorites, called chondrites, have strong similarities in relative chemical abundances to the sun and may be considered as « primary » bodies. While they often show evidence of element migration on a local (microscopic) scale and some recrystallization due to heating, they do not show the typical results of major planetary differentiation processes (melting, gross loss of volatiles, outgassing, large scale physical-chemical segregations). Some of the chondritic material has certainly resided and evolved within a planetary body. The chondrites, therefore, cannot be considered as cosmic virgins, but possibly as vestigial virgins. In general, these meteorites appear to preserve the characteristics of the earliest planetary material in the solar system. Some chondrites are found to contain chondrules and blebs of material rich in calcium and aluminum. Some of these inclusions are close in chemical composition to what would condense out of a hot cloud ($\sim$ 1800°K) of solar composition during cooling at a pressure of 10^{-3} bars. The major phases present in these so-called high temperature Ca-Al-inclusions are very similar to those calculated from thermodynamic considerations (see LORD, 1965; GROSSMAN, 1972) and are interpreted as direct condensates from a hot solar nebula. These inclusions often contain specks (or micron-size nuggets) of platinum group elements (WARK and LOVERING, 1977). Some of the platinum group elements, Hf, Os, W, are extremely refractory and may have been the earliest condensates to form. These could have been a soot which acted as the seed for later condensates. These chondrules and blebs of high temperature phases may represent early rain or hail (seeded, non-equilibrium droplets or aggregates which precipitated from a somewhat supercooled medium) from local portions of the solar nebula which became hot and were then subject to cooling. Many of the inclusions show clear textural evidence of having crystallized from a liquid (*cf.* BLANDER and FUCHS, 1975). The most spectacular examples of such inclusions are in the Allende meteorite which fell on 8 February 1969 near Pueblito de Allende, in the vicinity of Hidalgo del Parral, in Mexico.

The other class of meteorites may be called « secondary » objects. These include igneous rocks which are the obvious result of early planetary melting processes: the iron meteorites, stony-iron meteorites, and the achondrites (such as the eucrites, howardites). The achondrites are clearly

the result of crystallization of silicate melts and represent lavas or agglomerates of pieces of lavas. The iron and stony-iron meteorites also appear to be the result of local planetary melting with accumulation of metallic Fe-Ni and FeS in small pods or pools in the meteorite parent bodies. These secondary objects are of critical importance in understanding the processes which took place in the early stages of planetary evolution. They are distinct from the «primary» objects in being removed from the preceding nebular evolution by additional stages of planetary evolution. Our discussion will include isotopic observations on both «primary» and «secondary» meteorites, but with an emphasis on primary objects which contain high temperature Ca-Al rich inclusions which could represent the earliest condensates (MARVIN, WOOD and DICKEY, 1970; GROSSMAN, 1972).

RULES AND REGULARITIES OF ISOTOPES

The relative abundances of the isotopes of a given element may be changed by a variety of processes. Isotopes when charged may be strongly separated through the effects of electric and magnetic fields depending on the ratio of charge to mass and the kinetic energy of the particles. For uncharged particles, the separation is usually not highly specific and for small separations is dependent on the masses of the isotopic species. Such separation is possible as a result of chemical equilibrium between different phases (*e.g.*, solid-liquid-gas), from a host of kinetic effects and from settling in a gravitational potential. For kinetic effects the degree of separation depends on the extent to which the parent reservoir is depleted (*e.g.*, 10 % loss of matter, 90 % loss, *etc.*). There may of course be shifts in isotopic abundance due to the laboratory procedures which are used in chemical separation or in the actual measurements. If we consider the shifts in relative isotopic abundance to be a function of mass for small effects, then we obtain $N_{M+1}/N_M \approx (1+\alpha)(N_{M+1}/N_M)_0$, where α is the mass fractionation factor per mass unit difference and N_{M+1}/N_M is the ratio of the observed abundances relative to a standard (0). If this rule were followed then all isotopic variations would be as indicated by curve F-F in Figure 2. This abundance trend is commonly observed both due to laboratory effects and those which occur in nature. Deviations of one isotope from this trend are not plausibly due to mass fractionation and are called non-linear anomalies (see mass $M+1$ in Figure 2). These anomalies are presumed to be associated with nuclear effects. If, for example, only one isotope out of several is in excess relative to a fractionation trend, it may be considered to represent the addition of this isotope to the standard material. If it is deficient, then it may represent some special nuclear mechanism for the destruction of that isotope or the existence of material with a deficiency of that isotope relative to standard material. We may think of the average solar system abundances as representing mixtures of isotopes from different sources. If some exotic isotope is added to average solar material it will give an excess

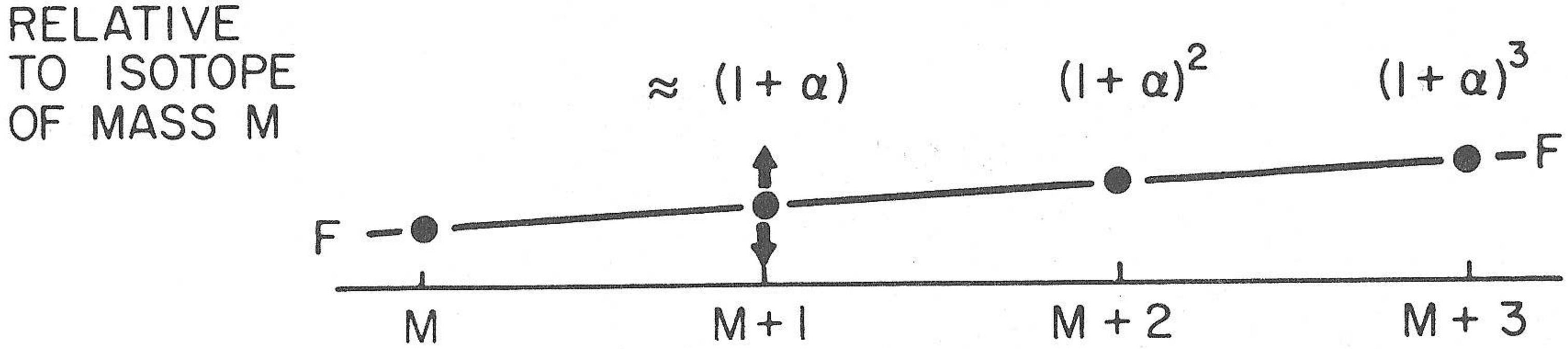


Fig. 2. Isotope fractionation pattern for a uniform mass fractionation factor $(1 + \alpha)$ per unit mass difference. For small fractionation effects, isotope ratios lie on a line FF. Deviations of an isotope $(M + 1)$ (see arrows) from the linear array defined by at least three other isotopes would correspond to an excess or deficiency assignable to this isotope and indicative of non-linear or nuclear effects.

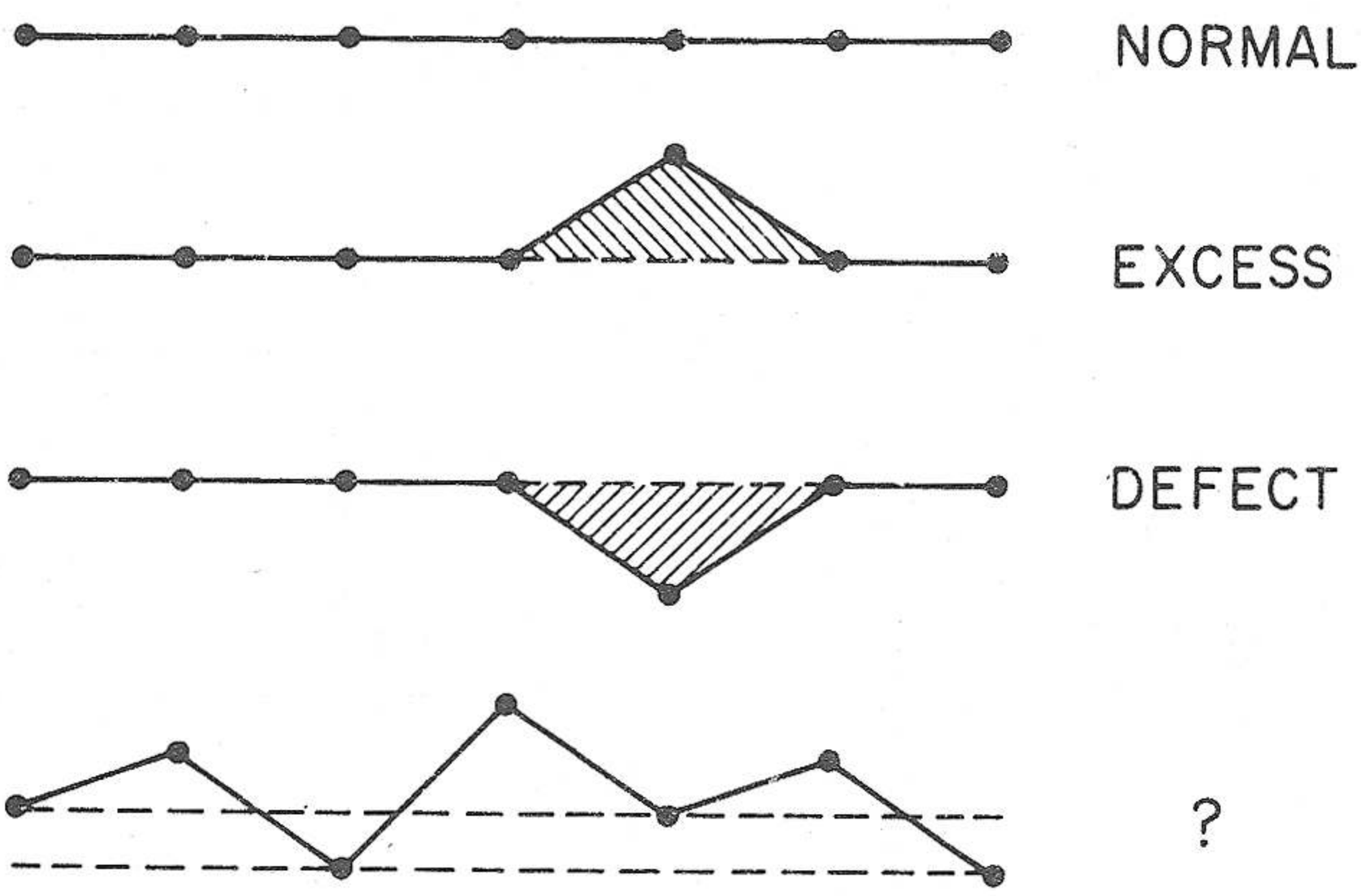


Fig. 3. Schematic patterns of isotopic anomalies relative to terrestrial abundances. The position of the dots in the top represents no fractional deviations from terrestrial values. An excess or a deficiency at a single isotope would be uniquely assignable. The second pattern corresponds to isotopes with terrestrial abundance but with one isotope in excess. For a more complex pattern (bottom) the identification of the isotopes which are anomalous depends on the normalization chosen, the baseline of the original unaltered abundances and possibly on other assumptions. In this case the identification of an anomaly is not unique.

of that isotope as indicated (Figure 3). If, however, the solar values were originally made up of some average value to which an isotope had to be added in order to make up the *present* average value, then an isotope deficiency would be observable at that mass for some samples and the deficiency would give a lower limit to the number of such nuclei which had to be added to the whole solar system. More generally, the solar system may have been heterogeneous in several isotopes of one element and it would no longer be possible to identify a simple excess or deficiency. We note that, for elements with only three naturally-occurring isotopes, unless the effects are much larger than possible instrumental mass fractionation factors, it is not possible to determine which isotope is actually anomalous. In the simplest case where the solar values for an element resulted from mixtures of material from two distinctive nucleosynthetic sources, we would expect variations in the isotopic abundances in mixtures of different proportions (see Figure 4).

As opposed to the more general isotopic anomalies, excesses are commonly observed for isotopes with long-lived radioactive parent isotopes which are known to exist today (*e.g.*, ^{40}Ar, ^{87}Sr, ^{206}Pb, ^{207}Pb, ^{208}Pb). Excesses which are not associated with existing parent isotopes may, however,

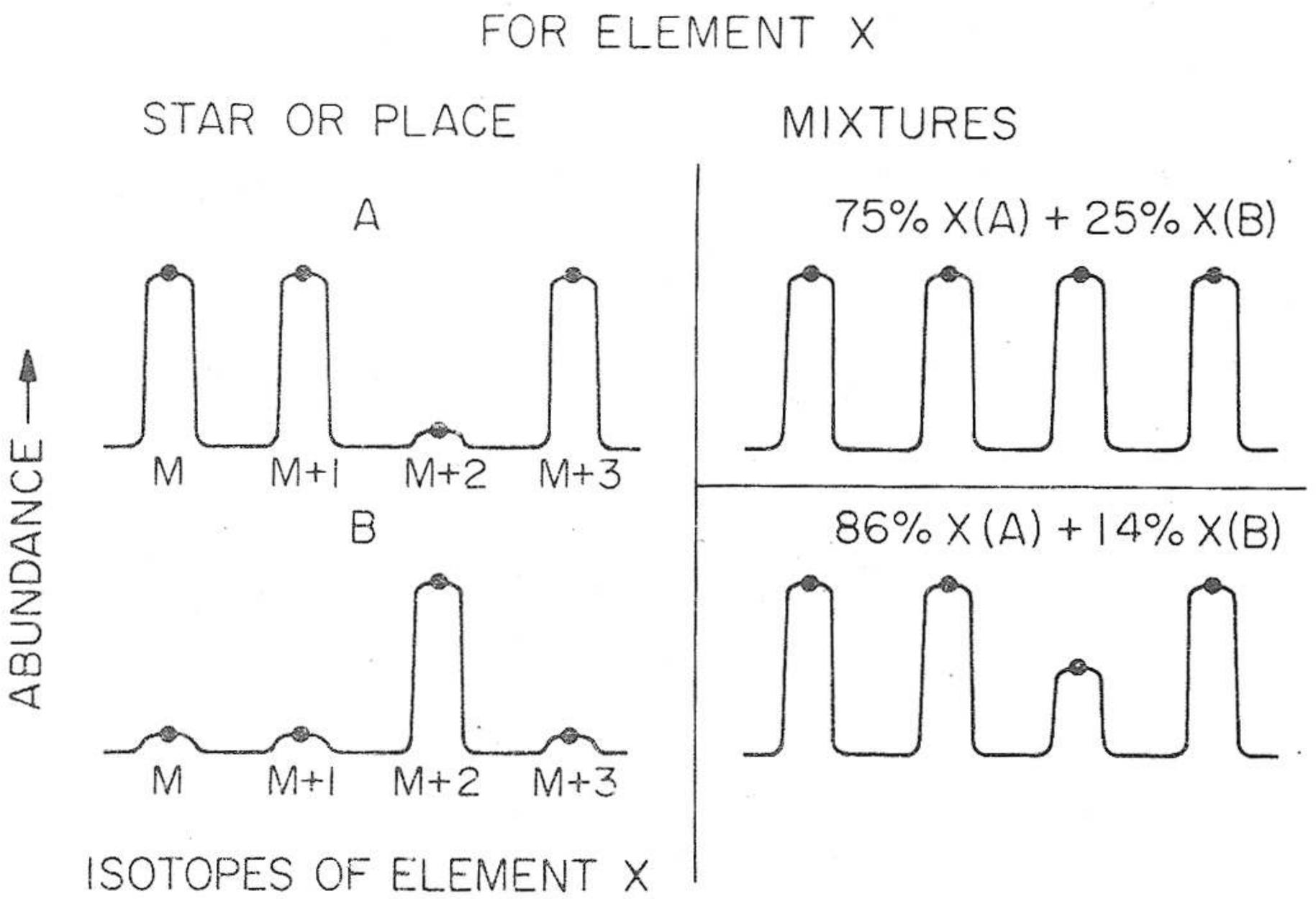


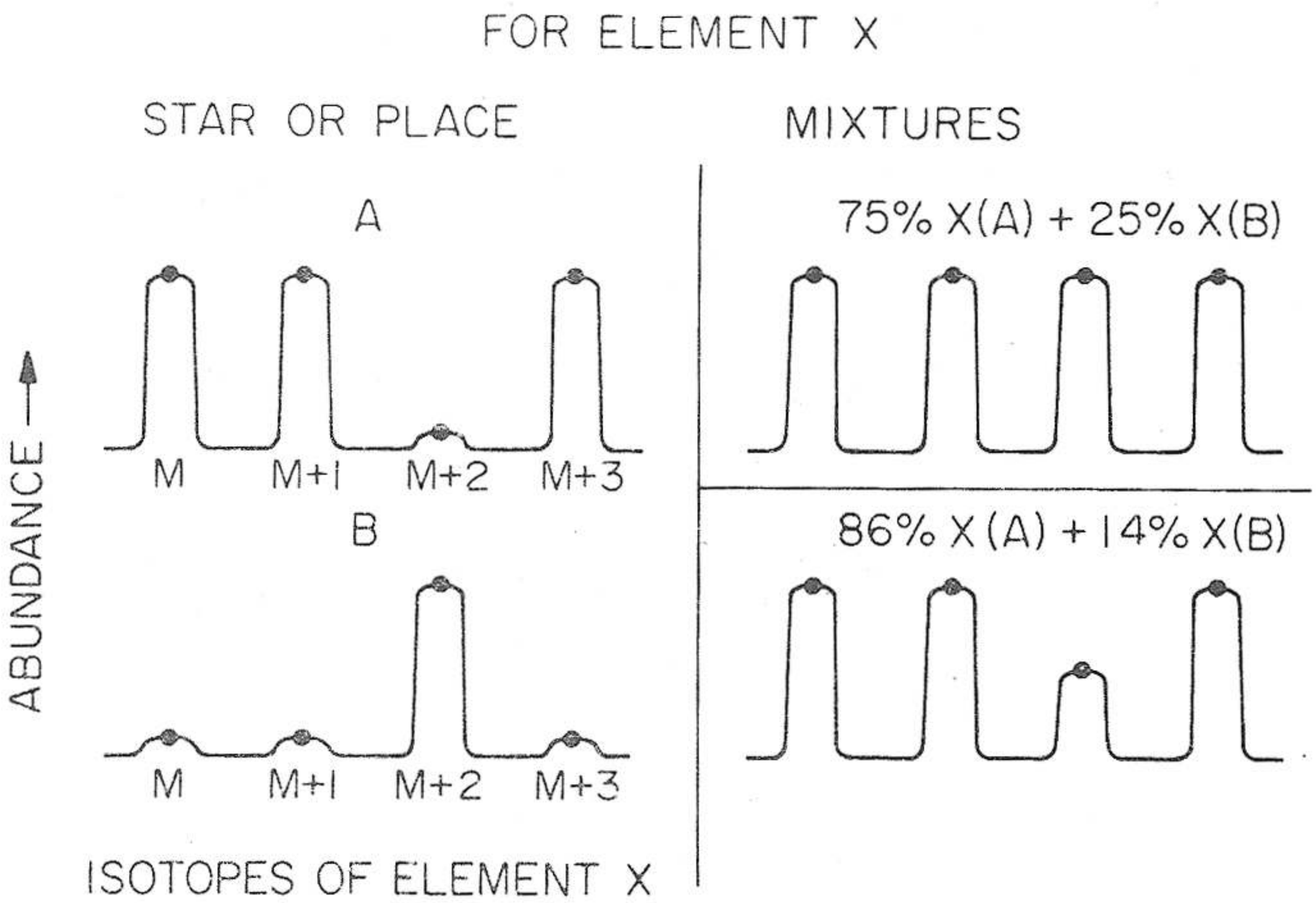
Fig. 4. Mixing of distinct isotopic patterns, produced in stars (A, B), to produce different observed mixtures (right). The existence of at least two contributing isotope patterns and the distinction at mass $M + 2$ would be deduced from the mixtures on the right, although the compositions A and B on the left would not be uniquely determinable without a reliable nuclear synthesis model.

result from the decay of parent radioactive isotopes which are now extinct (no longer found in nature). In order to attribute an excess of a given nuclide to an extinct radioactive nuclide, it is necessary to demonstrate that the excess nuclei have the same chemical distribution that the parent nuclide would have had under the circumstances when the object formed.

In addition to different nucleosynthetic mixes and the accumulation of decay products of radioactive nuclei, it is necessary to consider isotopic effects produced by nuclear reactions in dispersed media such as dust or gas by cosmic rays, and the reactions in condensed matter by cosmic rays and reactions from emitted high energy α's and fission of long-lived nuclei with the surrounding nuclei (*e.g.*, (α, n) and (n, γ)).

THE OXYGEN PROBLEM

Studies of different meteorites and of the earth and moon show that there exist small but distinct variations in the oxygen isotopic composition between solid planetary bodies in the solar system. This was first recognized by TAYLOR, DUKE, SILVER and EPSTEIN (1965) who found small variations of $^{18}O/^{16}O$ between meteoritic and terrestrial rock samples. CLAYTON, GROSSMAN and MAYEDA (1973) studied both the $^{18}O/^{16}O$ and $^{17}O/^{16}O$ abundances in inclusions in the Allende meteorite and showed that large variations existed in both ratios. These workers demonstrated that the correlation between the $^{18}O/^{16}O$ and $^{17}O/^{16}O$ effects was not due to mass dependent isotopic fractionation, as previously assu-



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med, but due to mixing of isotopically distinctive reservoirs. CLAYTON *et al.*, (1973) proposed a model in which interstellar dust grains containing essentially pure ^{16}O, without ^{18}O and ^{17}O, were mixed with normal solar system material. In addition to observing variations between different objects, CLAYTON, ONUMA, GROSSMAN and MAYEDA (1977) also reported large differences between coexisting mineral phases within individual inclusions from the Allende meteorite.

Let us consider the general patterns for the isotopic composition of oxygen using the three isotope correlation diagram for oxygen in which $^{17}O/^{16}O$ is plotted against $^{18}O/^{16}O$ (Figure 5a). The precise isotopic composition for oxygen in the sun, which is the main oxygen reservoir of the solar system, is not in fact known, nor are there values for the outer planets or their satellites. In Figure 5a we have arbitrarily placed the solar value at a distinct point labeled ⊙. Reliable data exist for the earth, the moon, a wide variety of meteorites (CLAYTON, ONUMA and MAYEDA, 1976; CLAYTON and MAYEDA, 1978) and for the atmosphere of Mars (NIER, McELROY and YUNG, 1976) and are indicated schematically in Figure 5a. This represents an approximation to the average value for the terrestrial planets. If a reservoir M were subject to isotopic fractionation, producing new complementary reservoirs M' and M'', then they would lie approximately on a line M' M M'' with a slope of 1/2 $(^{17}O/^{18}O)_M$ for small fractionations (Figure 5b). If all solar system reservoirs were simply the product of such processes and without extreme fractionations, then the mean solar system value (including ⊙) and all solar system samples would lie on such a line through the terrestrial value (⊕). The observations by CLAYTON *et al.*, (1973) showed that Allende inclusions were displaced from a fractionation line in the neighborhood of ⊕ in the direction of the origin by up to ~ 40 ‰ (4 %) of the length of the segment from the origin to ⊕. This clearly indicates the presence of at least two distinctive oxygen components. Samples of solar system material will thus cover an area in the oxygen isotope diagram (see polygon in Figure 5a) rather than a point or a line. It should be evident that if current solar system reservoirs are mixtures of two components coupled with fractionation, no strict statement can be made about the oxygen composition in the sun from samples of the terrestrial planets. Furthermore, comparisons of samples relative to an assumed « end member » may not fully reflect the displacement relative to the state of the solar system prior to admixing another component. It is possible to consider the observed oxygen isotopic shifts as the result of: (1) mixing an extraordinary material (E) consisting of nuclei containing essentially pure ^{16}O with normal uncontaminated (U) solar system material (Figure 6a); or (2) the result of mixing an extraordinary material (E *) containing essentially pure ^{17}O and ^{18}O with the presently observed terrestrial ratio with originally uncontaminated solar system material (U *), which is free of ^{18}O and ^{17}O (Figure 6b).

With regard to the model of addition of exotic pure ^{16}O to uncontaminated presolar material, it is possible that the E component is actually

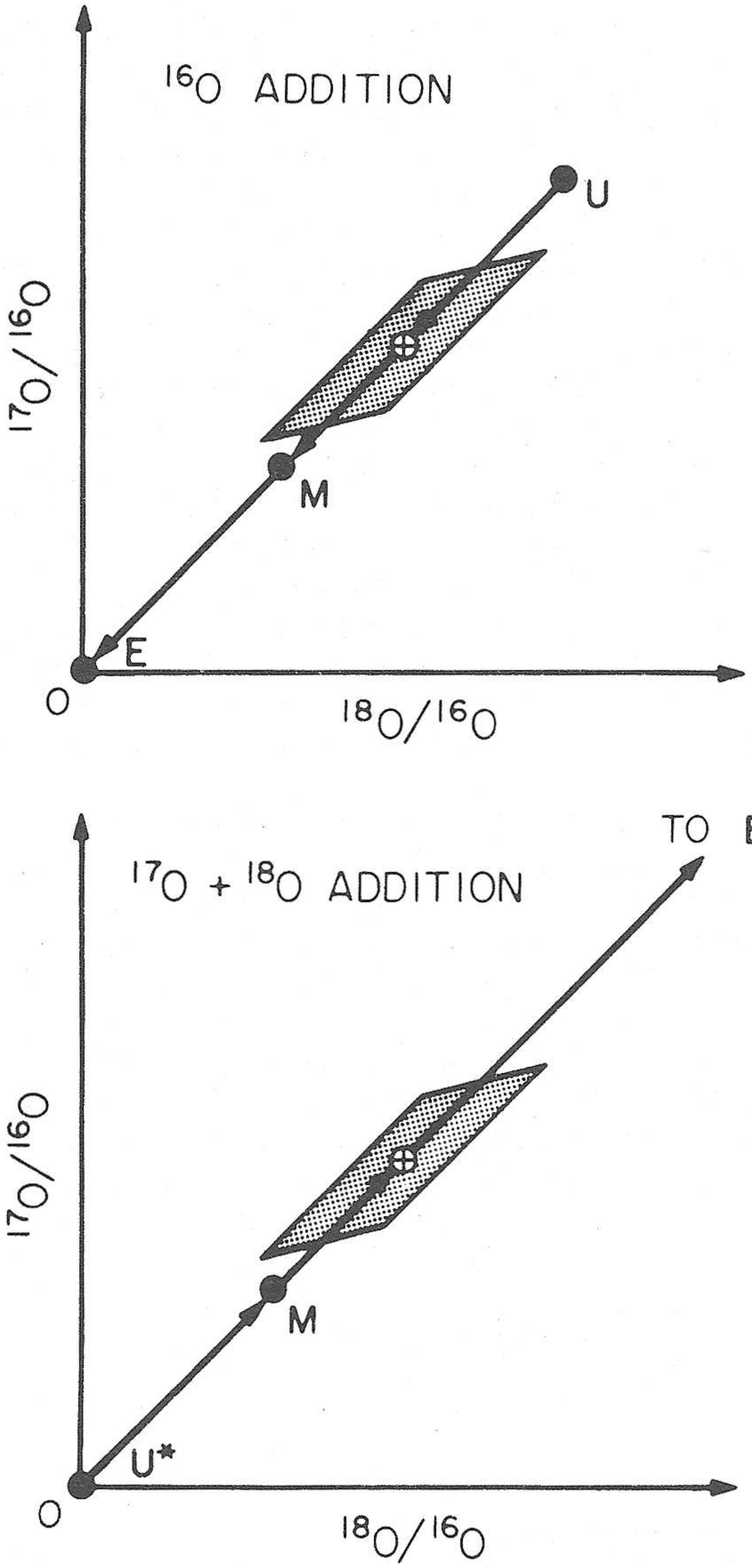

Fig. 6. (top) Schematic mixing diagram for uncontaminated (U) and extraordinary (E) reservoirs. The E reservoir is assumed to be ^{16}O-rich. Contaminating reservoir U with E causes the isotopic compositions to change in the direction of the arrows. (bottom) Same diagram for the U * and E * model where E * corresponds to the addition of ^{17}O and ^{18}O in a fixed proportion.

typical solar oxygen in which the ^{17}O and ^{18}O were destroyed by heavy proton bombardment by an active early sun (*cf.* LEE, 1978). For the model in which the uncontaminated solar system material U * contains essentially pure ^{16}O and the extraordinary contaminating material E * contains most of the ^{17}O and ^{18}O in the present solar system, the variations along the short arrow within the stippled region about ⊕ in Figure 6 are considered to be residual fluctuations of a process which is nearly complete. In contrast to the E model, the number of ^{18}O nuclei which must be added to produce the same isotopic shift for the E * model is rather small. If all of the ^{18}O (and ^{17}O) nuclei in a sample are from contamination with an E * reservoir, this constitutes only ~ 1/400 of all the oxygen nuclei. There is no apparent nucleosynthetic source of oxygen rich in ^{17}O and ^{18}O. In addition ^{17}O and ^{18}O seem to be widespread in the present interstellar clouds with an abundance roughly near terrestrial (within ~ 50 %, WANNIER, LUCAS, LINKE, ENCRENAZ, PENZIAS and WILSON, 1976). Thus there is no reason to believe that the proto-solar cloud was void of these species before the last injection. Furthermore, ^{18}O and ^{17}O are believed to be produced in two different sites rather than a single source (DEARBORN, TINSLEY and SCHRAMM, 1978). Therefore this model seems unlikely.

We consider here the model in which the total oxygen of a sample is a mixture of extraordinary oxygen (pure ^{16}O) from reservoir E and an ordinary uncontaminated solar system oxygen from reservoir U. The notation E follows BLACK's (1972) characterization of extra-solar ^{22}Ne. We assume that U contains all the oxygen isotopes and with normal $^{17}O/^{18}O$ (~ 0.18) but with $^{16}O/^{18}O$ not specified. Equations for $^{16}O/^{17}O$ may be written which are completely analogous to those given below for $^{16}O/^{18}O$. Consider a mixture (M) made of E and U and which has not been subject to isotope fractionation. Then the fractional deviation (Δ) in per mil of $^{16}O/^{18}O$ in M *relative to the uncontaminated solar material* U may be conveniently written as:

$$\Delta\,(^{16}O/^{18}O)_{MU} = \frac{[(^{16}O/^{18}O)_M - (^{16}O/^{18}O)_U] \times 10^3}{(^{16}O/^{18}O)_U}. \qquad (1)$$

It follows that

$$\Delta\,(^{16}O/^{18}O)_{MU} = (^{16}O_{EM}/^{16}O_{UM}) \times 10^3$$

where $^{16}O_{EM}$ and $^{16}O_{UM}$ are the numbers of extraordinary and of uncontaminated ^{16}O nuclei respectively in the mixture M. For measurements of M *relative to an arbitrary standard* S expressed as $\delta\,(^{16}O/^{18}O)_{MS}$ we have

$$\Delta\,(^{16}O/^{18}O)_{MU} = \frac{(^{16}O/^{18}O)_S}{(^{16}O/^{18}O)_U}\,[\delta\,(^{16}O/^{18}O)_{MS} - \delta\,(^{16}O/^{18}O)_{US}]. \qquad (2)$$

The usual form in which data are reported is $\delta\,(^{18}O/^{16}O)_{MS}$, which satisfies the identity:

$$[1 + \delta\,(^{18}O/^{16}O)_{MS} \times 10^{-3}]\,[1 + \delta\,(^{16}O/^{18}O)_{MS} \times 10^{-3}] = 1.$$

Note in equation (2) that $\Delta\,(^{16}O/^{18}O)_{MU}$ is not proportional to $\delta\,(^{16}O/^{18}O)_{MS}$ unless $\delta\,(^{16}O/^{18}O)_{US}$ is negligible. In general, $\Delta\,(^{16}O/^{18}O)_{MU}$ is not determined unless $(^{16}O/^{18}O)_{U}$ is known. The shift of ^{16}O relative to the « uncontaminated » value may in principle be quite large, although there is no evidence of wide differences (over 10 ‰) between different *large* planetary objects (CLAYTON and MAYEDA, 1978). However, the average solar system value for $\Delta\,(^{16}O/^{18}O)_{MU}$ may not be adequately approximated by $\delta\,(^{16}O/^{18}O)_{MS}$ for any of the usual standards (e.g., « *CCRS* », CLAYTON *et al.*, 1973).

It is not possible to determine reliably the contribution of exotic oxygen to the whole solar system. The variations of bulk meteorite samples found by CLAYTON and MAYEDA (1978) show a range of ~ 6 ‰. From the distinct oxygen isotopic composition of different classes of meteorites it is clear that the addition of an extraordinary component is not restricted to refractory inclusions in Allende but represents a wide spread phenomenon through at least all of the condensed material. The alternatives which appear to exist are the addition of ^{16}O (or of $^{18}O + {}^{17}O$) to the solar system: (a) in small amounts and added only to the material which makes up the terrestrial bodies, or (b) in large amounts to the whole solar system so as to significantly alter average uncontaminated solar system abundances. If meteorites provide good sampling of the solar system variability, then the amount of exotic oxygen added to the solar system must be at least 2×10^{28} grams of O_E. If the addition is only to the condensed material in the terrestrial planets, this will be reduced by a factor of $\sim 10^3$.

In the above discussion we have concentrated on the variations in oxygen isotopic composition on a gross scale. However, it is important to recognize that there exist large variations between coexisting mineral phases within a single morphologically well-defined inclusion. CLAYTON *et al.*, (1977) showed that different intergrown phases associated with early condensation (melilite, spinel, fassaite) showed different oxygen isotopic compositions in each individual inclusion. The spinel showed the largest amount of O_E component, fassaite the next largest, and the melilite showed no significant amount of an O_E component. For example, it was commonly found that $[\delta\,(^{18}O/^{16}O)_{spinel} - \delta\,(^{18}O/^{16}O)_{melilite}] \approx -30$ ‰. This variability suggested the presence of carrier grains which were derived from different nucleosynthetic sources and trapped in the inclusions. The question of whether each inclusion is an aggregate of grains from different sources (coarse stardust) or whether each inclusion and its constituent grains represent a single or homogenized source which was later altered is a fundamental issue which will be addressed in a later section. We note here that studies of other isotopic anomalies show the latter interpretation to be correct.

It is desirable also to explain other isotopic anomalies as part of the

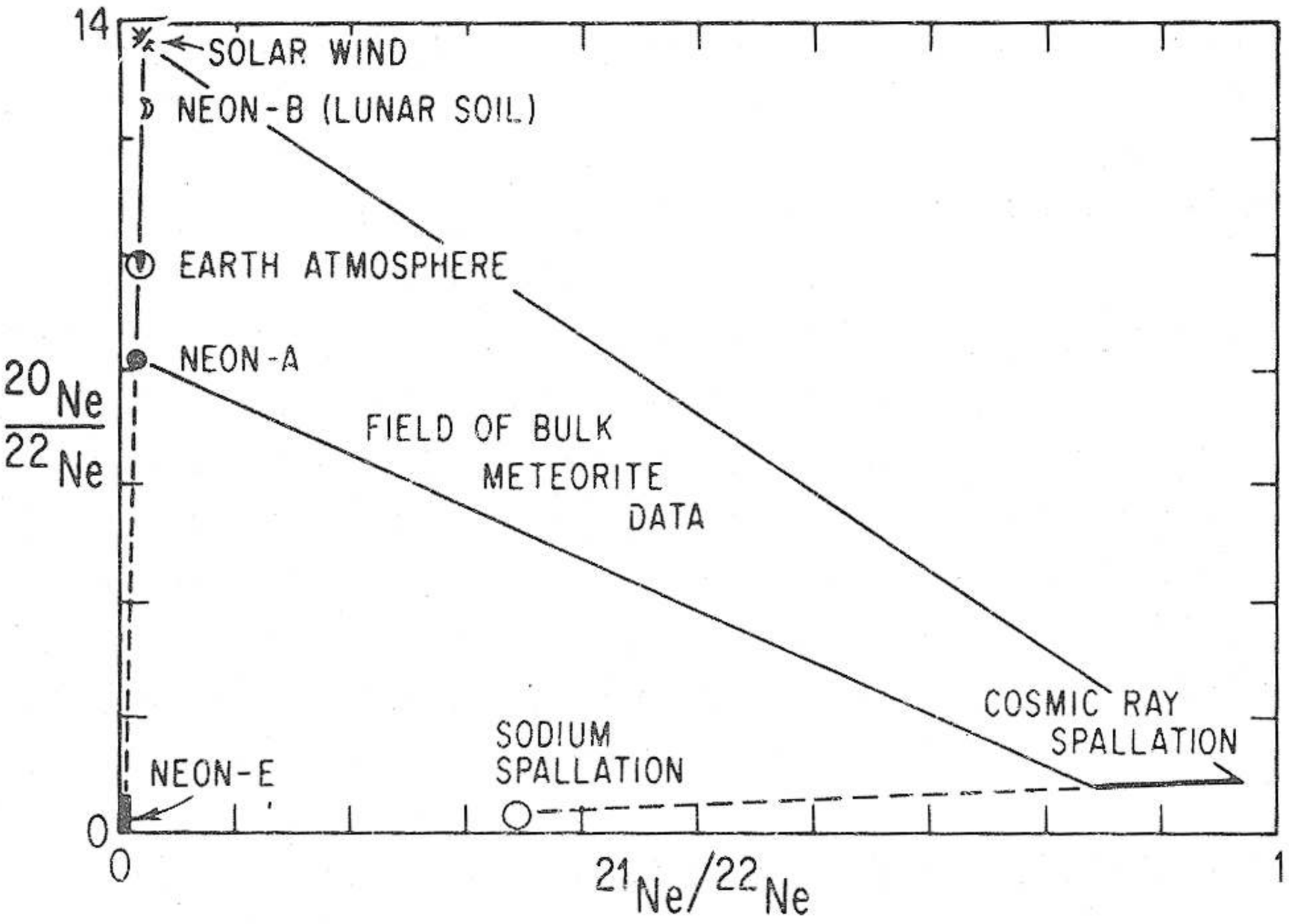

Fig. 7. Neon isotope compositions. Note the very low $^{21}Ne/^{22}Ne$ values if spallation effects are excluded. Components corresponding to different proportions of ^{20}Ne and ^{22}Ne plot very near the ordinate and exhibit extreme variations. The limits on the Ne-E component found in meteorites are shown by the shaded area near the origin. The earth's atmosphere and most meteorites are enriched in ^{22}Ne as compared to the sun (from direct measurements on solar wind Ne). The lunar Ne which is from implanted solar wind on lunar rocks is close to solar wind and altered by fractionation processes.

same addition phenomenon as found for oxygen. As first pointed out by BLACK (1972) and confirmed and refined by EBERHARDT (1974,1978), a component of possibly pure ^{22}Ne (called Ne E) with $^{20}Ne/^{22}Ne < 1.3$ and $^{21}Ne/^{22}Ne < 0.015$ is present in the solar system (*cf.* Figure 7). From measurements of solar wind (GEISS, BUEHLER, CERUTTI, EBERHARDT and FILLEUX, 1972) it is found that ^{22}Ne is present in the surface of the sun with an abundance of $^{20}Ne/^{22}Ne \simeq 13.7$, while the terrestrial value of $(^{20}Ne/^{22}Ne)_{\oplus} = 9.8$. The sun appears to be rich in ^{20}Ne while the planets and meteorites are relatively rich in ^{22}Ne. The high ^{22}Ne abundance relative to ^{20}Ne in planetary gases as compared with the solar wind suggests that the added extraordinary Ne-E component is enriched in the dusty and condensible material making up the terrestrial planets. This suggests that a quantitative correlation should exist between Ne_E and O_E. This is particularly appealing since ^{16}O and ^{20}Ne are produced in explosive C burning. No such correlation has been found, possibly due to the fact that there are no obvious mechanisms which would concentrate Ne in easily condensible materials or solids such as dust grains or in crystals which are high temperature condensates. An estimate of exotic Ne contributions relative to exotic oxygen assuming that only the terrestrial type materials were contaminated is thus not possible because Ne is severely depleted in condensed objects.

It is possible that the variations in oxygen which are observed are not due to the addition of extraordinary material to the solar system, but rather due to differences in O between gas and dust. In this case the O isotopic variation is the result of incomplete exchange or mixing between the reservoirs of distinct isotopic character, *e.g.*, gas and dust, in the proto-solar-cloud. It is most consistent to associate the pure(r) ^{16}O component with the dust phase. The sizes of the two O reservoirs can be estimated using the present solar abundance if all the easily condensible elements such as Ca, Al, Mg, Si, Fe, etc., were in the dust. The fraction of total oxygen initially in the dust must be approximately $[(Mg + Fe + Si)/O]_{\odot} \sim 0.15$ for any reasonable mineralogical composition of the dust. So the solar system oxygen was originally resident in two reservoirs which are different in size by only a factor of 7. Presumably, evaporation of the dust and gas-dust reactions would be the processes responsible for isotopic exchange between such reservoirs. A model of this type cannot explain the presence of short-lived nuclides, nor can it explain anomalies in refractory elements if the dust is treated as a homogeneous component. Therefore this model may only be considered as a partial source of isotopic heterogeneity in a stratified solar nebula. If various objects in the solar system are formed as mixtures of refractory materials residing in grains and of more « volatile » material residing in the gas, then we would expect the size of isotopic anomalies to correlate with the « refractory »/« volatile » ratios. For the oxygen data such a progression of isotopic effects is not apparent since the most volatile materials (carbonaceous chondrites) show intermediate effects between refractory Allende inclusions and ordinary meteorites (CLAYTON *et al.*, 1976; CLAYTON and MAYEDA, 1978). This indicates that for any model we require locally distinct vaporization-condensation histories and preservation of isotopic anomalies in parcels of gas during the high temperature stage of the nebula as well as the addition to both gas and dust of freshly synthesized nuclides.

FUN INCLUSIONS

Magnesium and Oxygen. One of the difficulties in understanding the observed isotopic anomalies has been the apparent lack of anomalies in refractory elements and the lack of correlation between effects in different elements except for inferred parent-daughter pairs. For some time, efforts to find a correlation between oxygen anomalies and the magnesium isotopic composition yielded no positive results (LEE and PAPANASTASSIOU, 1974). In most cases the Mg isotopic composition was found to be the same (and indistinguishable from terrestrial) in both spinel, pyroxene, and melilite from the same Ca-Al rich inclusions where the oxygen was highly anomalous for the spinel and pyroxene and nearly normal for the melilite. This could be due to an absence of any anomalies in Mg or to a much smaller exotic Mg component as compared with O_E. Correlations have been established for isotopic anomalies between the heavy rare gases (Kr, Xe), but they are not directly associated with any mineral phase nor

are they shown to be correlated with oxygen. Correlations in isotopic anomalies for different nuclear species are extremely important as they should provide a signature of the nucleosynthetic processes which caused them.

The first samples to exhibit peculiar isotopic compositions for several elements were found in two inclusions, C-1 and EK-1-4-1. These inclusions are Ca-Al rich chondrules from the Allende meteorite. They do not appear distinctive in terms of mineral composition or morphology from the typical high temperature Ca-Al rich inclusions. The original identification of the special nature of these samples came from the observation that they had (1) Mg isotopic compositions which were widely displaced from terrestrial values and could not be explained by decay of ^{26}Al, and (2) O isotopic compositions which did not lie on either the terrestrial fractionation line or on the $O_E - O_N$ line [LEE and PAPANASTASSIOU, 1974; LEE, PAPANASTASSIOU and WASSERBURG (LPW), 1976; CLAYTON and MAYEDA, 1977; WASSERBURG, LEE and PAPANASTASSIOU (WLP), 1977]. The oxygen data showed that the coexisting phases in each inclusion had different isotopic compositions and that these could be explained either by the presence of a third component (different from O_E or O_N) or as the result of isotopic fractionation of material which was a mixture of O_E and O_N. The Mg data showed that all of the coexisting phases (both for macroscopic and microscopic samples) had the same Mg isotopic composition and that the isotopes were regularly enriched with increasing mass number. The enrichments were almost precisely what would obtain if they were the result of isotopic fractionation (see Figure 8). However, there were small, significant departures from any simple mass fractionation law which indicated that ^{26}Mg was deficient if ^{24}Mg and ^{25}Mg were originally terrestrial in abundance but subsequently shifted by fractionation. (Note that it is not possible in this case of three isotopes to distinguish between ^{24}Mg deficit, ^{25}Mg excess, or ^{26}Mg deficit.) Except for these isotopic peculiarities, the inclusions C-1 and EK-1-4-1 appeared to be indistinguishable, based on mineralogical and textural relationships, from the typical type B Ca-Al inclusions (GROSSMAN, 1975). The Mg data could be explained by a model in which each inclusion was made of peculiar material that had been isotopically homogenized in the inclusion. This could be produced by melting of an aggregate of isotopically heterogeneous matter (say, an interstellar dust ball) or by condensation of a liquid droplet from a supercooled homogeneous gas of isotopically peculiar matter. The crystals which then grew in the inclusion (up to 3 mm) would each have the same peculiar Mg isotopic composition. The isotopically peculiar material must have been subject to strong isotopic fractionation and contained a small component of exotic Mg. While the oxygen data for the bulk EK-1-4-1 and C-1 could be similarly interpreted (fractionation of material which initially was comprised of a given $O_E - O_N$ mix), this model does not explain the differences in oxygen isotopic composition between the coexisting phases spinel, pyroxene, and melilite (see Figure 9).

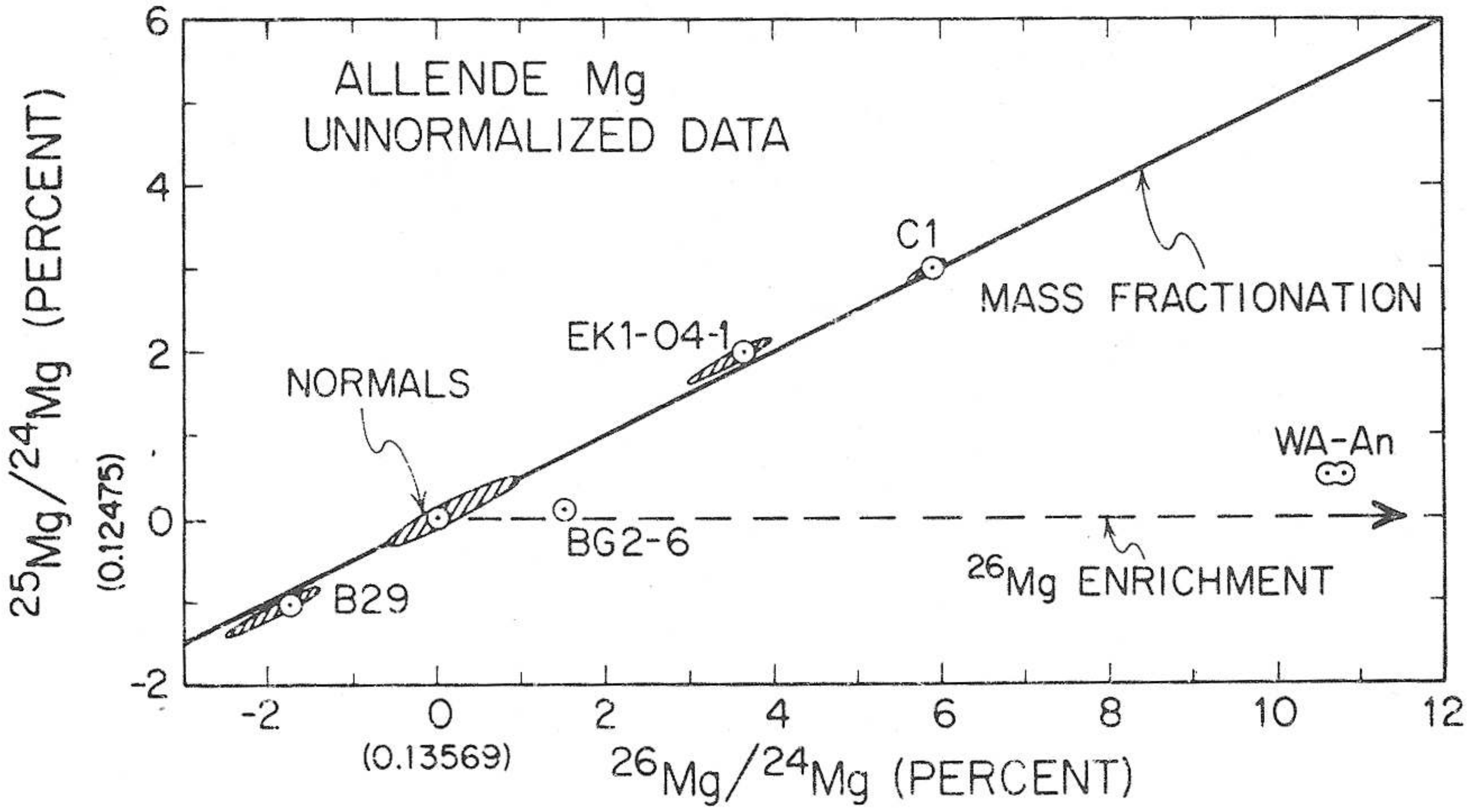

Fig. 8. Mass fractionation effects for Mg. We show raw measured data as percent deviations from the mean $^{25}Mg/^{24}Mg$ and $^{26}Mg/^{24}Mg$ for normal Mg (terrestrial, lunar, and chondritic). Shaded areas correspond to the observed maximum range of isotopic fractionation for each type of sample. The precision of the data is much better than the schematic display of the data range. FUN samples C-1 and EK-1-4-1 plot very near the expected correlation line for mass fractionation. Small but well defined deviations for these samples on the upper side of the mass fractionation line are evidence for superimposed nuclear anomalies or a more complex law of mass fractionation. Sample B-29, a fine-grained Ca-Al-rich aggregate from Allende, shows a complementary fractionation pattern (depleted heavier isotopes). Effects due to pure ^{26}Mg enrichment move points to the right along a horizontal line (*e.g.*, WA-An, anorthite from inclusion WA). [After WLP, 1977].

The interpretation of the Mg and O data by both WLP (1977) and CLAYTON and MAYEDA (1977) is that these Ca-Al rich chondrules were formed from a local parcel of gas and dust in the solar nebula which contained some exotic nuclei or nuclei with abundances somewhat different from the terrestrial values. The formation process involved large isotopic fractionation so that all of the primary mineral phases which grew in the chondrules were isotopically uniform but anomalous and fractionated. In a later stage, the minerals melilite and to some extent pyroxene were susceptible to a « back reaction » with cooler portions of the solar nebula, rich in volatiles and « normal » oxygen, thus causing a large shift in the oxygen composition of the melilite and only small shifts in the other phases. As Mg would be depleted in a volatile rich, oxygen rich (refractory poor) medium, the melilites would maintain their original peculiar Mg isotopic composition but shift in oxygen composition. This model is *ad hoc* but constitutes an explanation coupling the high degree of alteration readily observed in the melilites and their nearly normal oxygen composition. The pyroxenes only show minor alteration. This interpretation follows the suggestion for oxygen proposed by BLANDER and FUCHS (1975). The

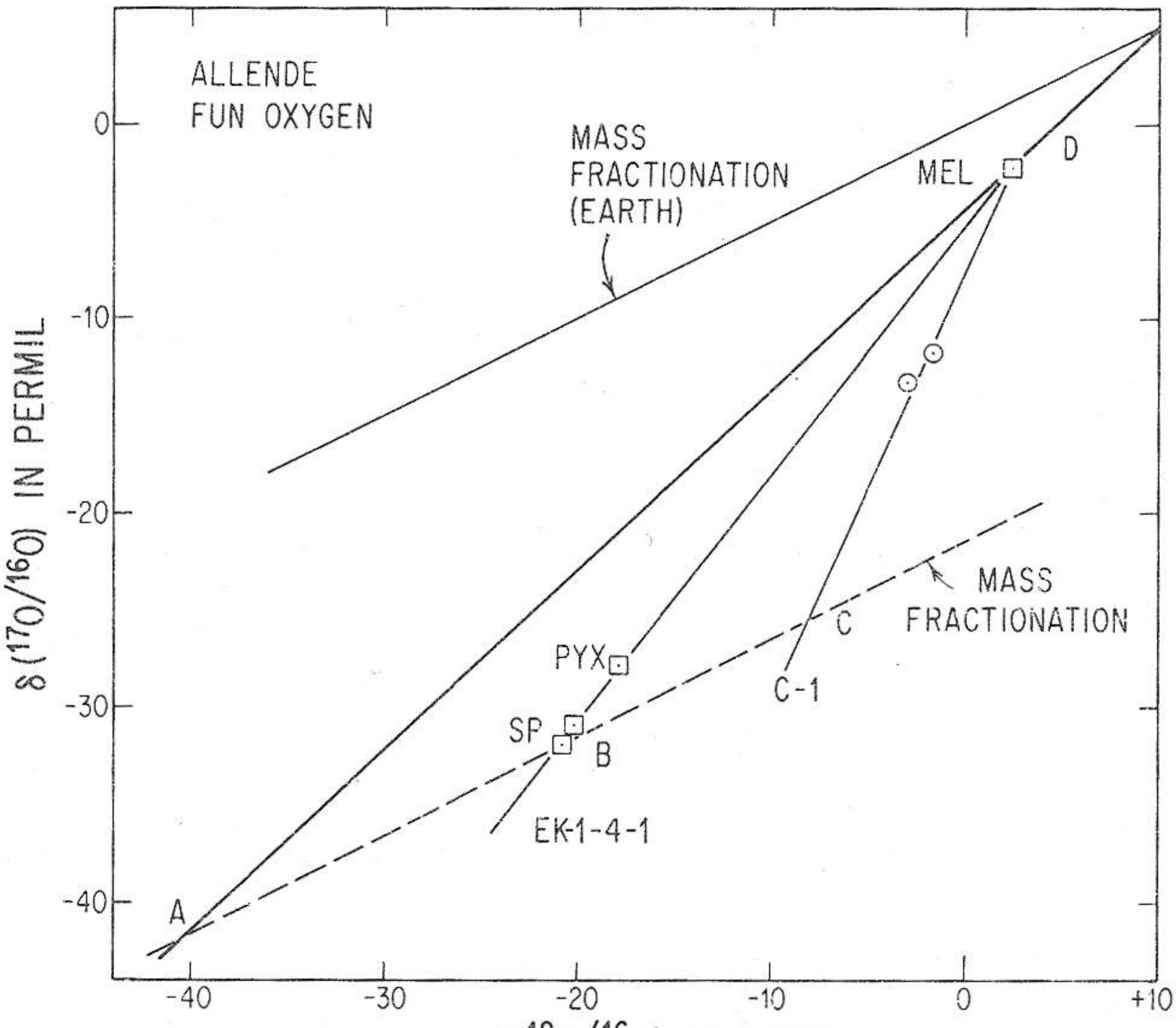

Fig. 9. Oxygen isotope compositions for FUN inclusions EK-1-4-1 and C-1. Data for mineral separates of EK-1-4-1 (squares) are shown for spinel (SP), pyroxene (PYX), and melilite (MEL) and define line BD. Data on samples of the total inclusion C-1 are shown as circles. The data for both inclusions are far off line AD defined by oxygen measurements of total inclusions and mineral separates from all other Ca-Al-rich Allende inclusions. Note that EK-1-4-1 melilite is close to the terrestrial data line and is consistent whith a model in which FUN oxygen with a composition near point B has largely exchanged with oxygen of normal composition in the vicinity of D. If one assumes that the oxygen composition in EK-1-4-1 prior to exchange with normal oxygen is at point B (SP + PYX) then this oxygen composition could have been obtained from an initial composition A on the O_E–O_N mixing line (line AD), where line AB, with slope 1/2 corresponding to mass fractionation, was simply drawn through point B. Displacement of FUN inclusions off O_E-O_N line could also reflect distinct extraordinary oxygen compositions. The interpretation of displacement off line AD as due to mass fractionation was dependent on the observation of large fractionation effects for Mg in the same samples. [after CLAYTON and MAYEDA, 1977].

fine-grained alteration products observed in the Ca-Al rich inclusions have not been studied extensively, but they are rich in Na, Cl, and other volatile elements and contain phases which could only form at much lower temperatures than spinel, melilite, and Ca-Mg- Ti-rich pyroxenes. These alteration products are seen to permeate the melilite crystals in veins and throughout their volume.

The general character of the isotopic shifts originally found for Mg and O for C-1 and EK-1-4-1 suggests two distinct effects: (1) isotopic fractionation (F), and (2) unidentified nuclear (UN) effects. This correlation

caused WLP (1977) to define these as FUN anomalies. As will be shown later, the existence of «nuclear» effects in many elements is apparently closely tied to the presence of mass fractionation. Subsequent to the identification of these two FUN samples based on the Mg and O observations, it was discovered that a host of other refractory elements were isotopically peculiar in these same inclusions. The extent to which the non-linear or nuclear effects are related to the fractionation observed in O and Mg is not clear. Clearly some elements show no fractionation but have nuclear anomalies. In all cases some care must be taken in identifying deviations from a particular fractionation law as reflecting true nuclear anomalies. For most samples which show small fractionation this is not important but in some critical cases both the identification and magnitude of a nuclear anomaly will depend on the fractionation law that is assumed. There is at present no satisfactory interpretation for the apparent correlation between fractionation and nuclear anomalies. Having identified a correlation between the peculiar isotopic patterns for both O and Mg in the two FUN inclusions, it was of importance to ascertain whether any other elements exhibited peculiar isotopic patterns. This search proved difficult but successful and the resulting anomalies which have now been established are discussed below in order of increasing atomic number. The data on Ca, Ba, and Nd were first reported at the 21st R. A. Welch Foundation Conference on Chemical Research in November, 1977, in Houston (WASSERBURG, 1978).

Aluminum 26. A search was conducted for evidence of ^{26}Al in sample C-1 by ESAT, LEE, PAPANASTASSIOU and WASSERBURG (1978). No data have been obtained on EK-1-4-1 because of the minute amount of material which remains. These workers report that there is a suggestion of ^{26}Mg excess resulting from ^{26}Al decay in plagioclase crystals ($CaAl_2Si_2O_8$) in C-1, but with a $^{26}Al/^{27}Al$ ratio which is a factor of seven less than found in typical Ca-Al rich inclusions in Allende (see general discussion on time scales). They further report the presence of some Mg of terrestrial composition in impure plagioclase and attribute this to a back reaction process as outlined by WLP (1977). The effects of chemical alteration subsequent to the formation of high temperature condensates are strongly emphasized by these authors. The strongest conclusion is that only a small amount of ^{26}Al, and hence fewer late stage nucleosynthetic products, were included in the C-1 inclusion as compared with other materials (see section on time scales for a fuller discussion of ^{26}Al).

Silicon. The presence of large isotopic fractionation effects as observed for Mg and inferred for O were substantiated by YEH and EPSTEIN (1978) and CLAYTON, MAYEDA and EPSTEIN (1978), who found that Si was also strongly fractionated in these two inclusions. The results show clear evidence of isotopic fractionation with the heavier isotopes being enhanced similar to what was found for Mg (see Figure 10). There is a close relationship between the degree of fractionation for Si and Mg. Sample C-1 is the most fractionated and EK-1-4-1 is less fractionated. There appear to be some small departures from a simple fractionation behavior for sample

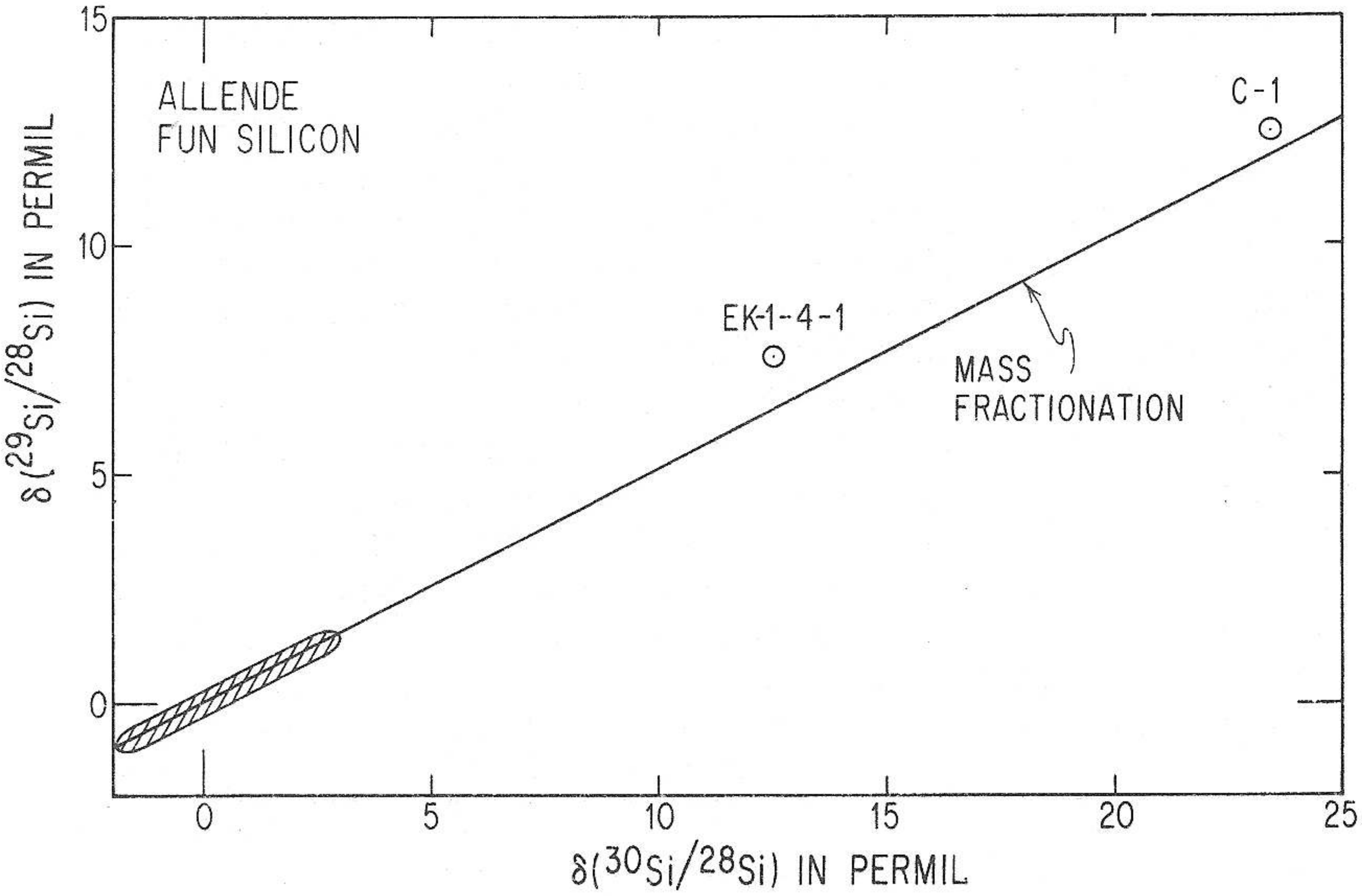

Fig. 10. Silicon composition for FUN inclusions EK-1-4-1 and C-1. The hatchured area shows the extent of variations in Si isotope composition along the calculated mass fractionation line for meteorites and non-FUN Allende inclusions. Large displacement of the FUN samples along the mass fractionation line yields Si fractionation effects which are comparable to those observed for Mg in these samples. Deviations off the Si fractionation line are not well resolved analytically but suggest the presence of nuclear effects (relative deficiency of ^{30}Si or excess of ^{29}Si). [after YEH and EPSTEIN, 1978 and CLAYTON *et al.*, 1978].

EK-1-4-1, but there is no high precision, reliable Si data to justify a firm conclusion regarding the presence of a peculiar nuclear component in Si.

Calcium. The chart of the nuclides in the neighborhood of Ca is shown in Figure 11. The abundances given in the chart are not the percent abundance of each isotope as is usually given but the « solar » abundance of each nuclear species (see CAMERON, 1973a). This tabulation permits a direct comparison of observed isotopic effects with the number of nuclei which must be altered, relative to the « solar » values with Si = 10^6. All variations in isotopic abundances are given relative to terrestrial standards and are expressed in fractional deviations either in parts in 10^3 (permil) called δ units or in parts in 10^4 called ε units. Note that the relative abundances of the Ca isotopes range over a factor of $\sim 3 \times 10^4$. The nucleosynthesis of the Ca isotopes is not well understood, but they are produced by distinctive processes (explosive oxygen and silicon burning, neutron capture on seed nuclei; WOOSLEY, ARNETT and CLAYTON, 1973 and HOWARD, ARNETT, CLAYTON and WOOSLEY, 1972). As a result of the wide range in relative abundances and nucleosynthetic processes, Ca was considered as a key element for study. From a technical point of view

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Figure 11

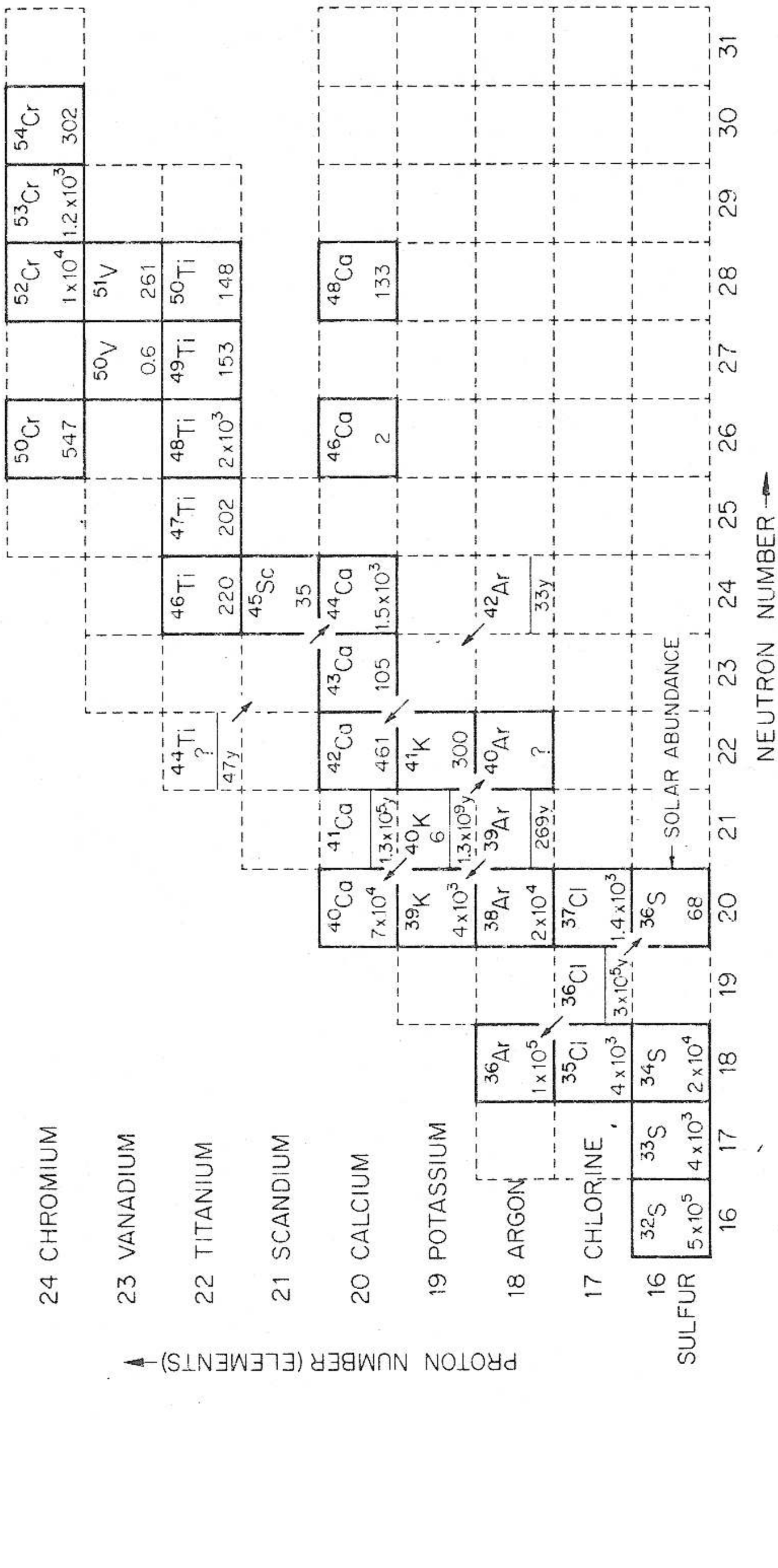

Fig. 11. Chart of the nuclides in the vicinity of Ca. Dashed lines indicate unstable nuclides. The entries in each square include the solar abundance of each nuclide from CAMERON (1973a) using the $^{28}Si = 10^6$ normalization. Half-lives are given for the longerlived radioactive nuclides. Note the existence of very neutron rich Ca isotopes as well as the large abundance difference of the Ca isotopes (7×10^4 to 2).

the measurement of relative Ca isotopic abundances is made difficult by both the wide range in these abundances and the large mass difference which may result in large instrumental mass fractionation. In the case of large fractionation the particular « law » used to reduce the data can affect both the quantitative and qualitative nature of « non-linear » isotopic effects. Most samples of Ca in nature show only small fractionation effects. A thorough discussion of both procedures and data is given by RUSSELL, PAPANASTASSIOU, and TOMBRELLO (1978).

The results for C-1 and EK-1-4-1 are shown in Figure 12 (LPW, 1978).

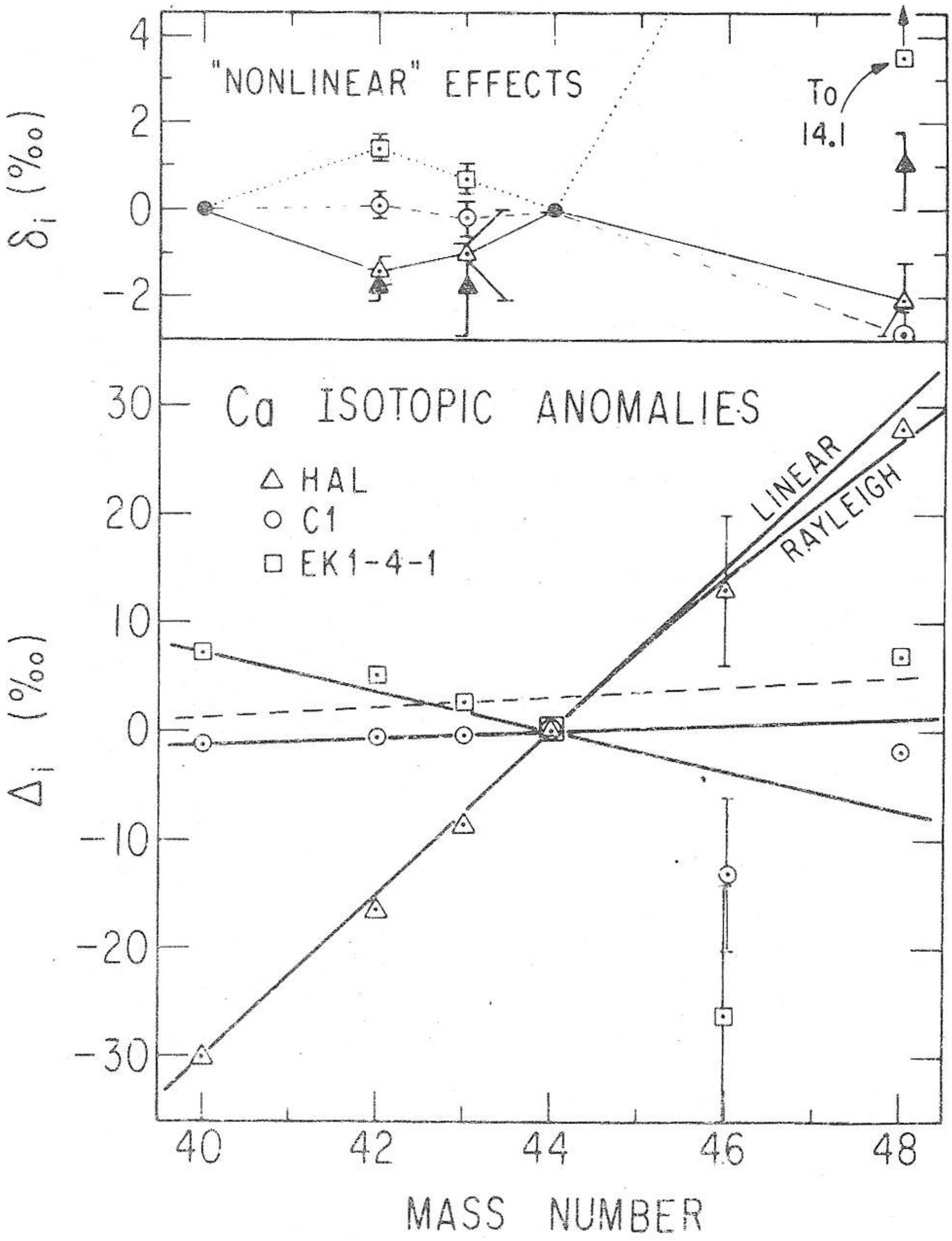

Fig. 12. Fractional differences of Ca isotopes relative to normal ($\delta = 0$ line) normalized to ^{40}Ca and ^{44}Ca. The upper graph shows non-linear effects after correction for mass fractionation. Sample EK-1-4-1 shows excesses in ^{42}Ca and ^{48}Ca (off-scale) while sample C-1 shows a ^{48}Ca deficiency. The HAL sample shows only small non-linear effects at ^{42}Ca but effects at ^{48}Ca are not clearly resolved due to uncertainties as to which fractionation law is applicable. The lower graph shows ratios relative to ^{44}Ca only which demonstrate the large fractionation observed for Ca isotopes in the HAL, hibonite rich inclusion. Linear and Rayleigh fractionation laws (for HAL) are shown. [after LPW, 1978, and LEE *et al.*, 1979].

We have also included results from a third FUN inclusion which was recently discovered (LEE, RUSSELL and WASSERBURG, 1979). The experimental errors for ^{46}Ca are $\pm \sim$ 10-20 ‰ and have been omitted from the figure for clarity. Assuming that the ^{40}Ca and ^{44}Ca are unaffected by nuclear effects, neither of these samples appears to be seriously fractionated as determined by measurements using a double spike (LPW, 1978). Sample C-1 shows all isotopes to be normal within error except for a depletion in ^{48}Ca by about 3 ‰. It is surprising that ^{46}Ca shows no large anomalies since it is only 1/60th as abundant as ^{48}Ca. Shortage of the same number of nuclei at ^{46}Ca as is observed at ^{48}Ca would cause a 180 ‰ effect. As all other isotopes appear normal in C-1, it is plausible to attribute the pattern to a deficiency in ^{48}Ca. This indicates that the ^{48}Ca which was last injected to produce the average solar system value must have been generated by a rather special mechanism which favored this nucleus or that ^{48}Ca was preferentially destroyed in a parcel of terrestrial type Ca.

The data on EK-1-4-1 are shown on the same figure and indicate very large relative enhancements at ^{48}Ca of 14 ‰ and at ^{42}Ca of 1.6 ‰. The EK-1-4-1 results appear to indicate addition of ^{48}Ca and ^{42}Ca to average solar material with the fractional effects being ten times larger at 48 relative to 42. The Ca data on C-1 and EK-1-4-1 taken together suggest that solar system Ca is the result of the incomplete mixing between the uncontaminated bulk material of the solar nebula which was substantially deficient in ^{48}Ca and slightly deficient in ^{42}Ca relative to normal and a special nuclear component consisting of ^{48}Ca and ^{42}Ca in the ratio of 2.5 to 1 by number. In this interpretation normal Ca is also a mixture and the deficiency in ^{48}Ca implies that the special nuclear component accounts for at least 3×10^{-3} of the ^{48}Ca and 5×10^{-6} of the total Ca presently in the solar system.

Strontium. The Sr isotopic variations in C-1 and EK-1-4-1 are shown in Figure 13 with isotopes 86 and 88 used for normalization. Both samples show a deficit of the rare isotope ^{84}Sr which is produced solely by the *« p »* process (PAPANASTASSIOU and WASSERBURG, 1978). It would be simplest to assign the effect to a shortage in ^{84}Sr *(p)* or an enhancement in $^{86, 87, 88}Sr$ which are all dominantly *« s »* products. However, this is not in accord with the evidence in Ba, Nd, and Sm in these samples. In particular, EK-1-4-1 shows excesses in the *r*-process isotopes for Ba, Nd, and Sm, and a *p*-process excess in Sm but null *p* isotope effects in Ba; C-1 shows no effects for Ba and Nd (except for a small ^{135}Ba deficiency) and a *p*-excess for Sm.

The assignment of the Sr isotopic anomaly to ^{84}Sr is not unique. A check can be made using ^{87}Sr, the only other isotope not used for normalization. The isotope ^{87}Sr is produced dominantly by the *« s »* process and partly by the decay of ^{87}Rb so that it is not possible to assign a very precise normal value to its abundance and to establish whether further nuclear effects are present in this isotope. ^{87}Sr increases $\sim$ 0.005 % per 10^6 y due to radioactive decay of ^{87}Rb in a mix of solar proportions. The observed values of $^{87}Sr/^{86}Sr$ when corrected for ^{87}Rb decay for 4.6 AE lie close to the

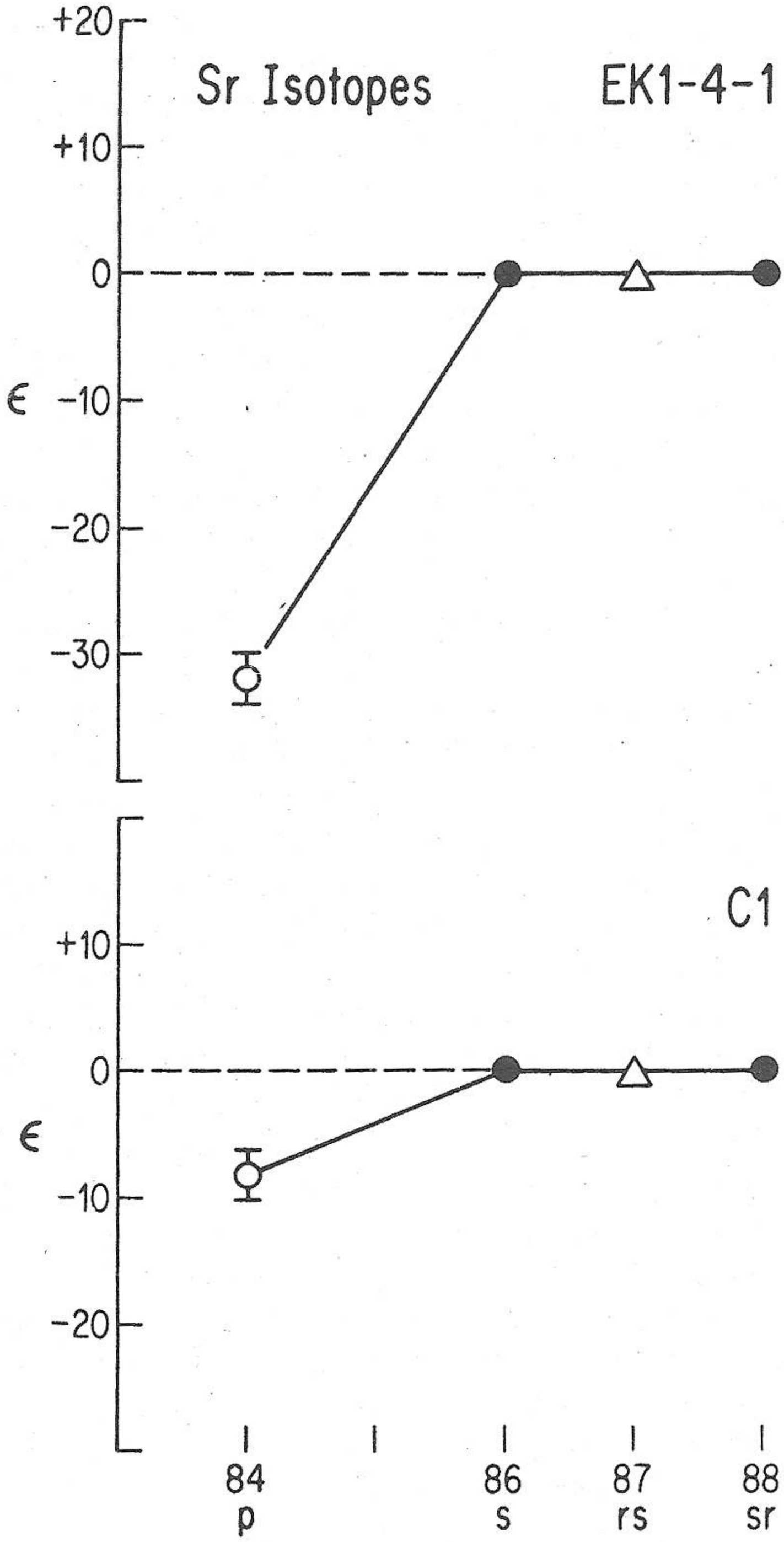

Fig. 13. Sr isotope composition for FUN inclusions. In this and subsequent graphs the pair of isotopes assumed to have normal composition are shown as full circles; the rest of the isotopes are shown as open circles. The units are in parts in 10^4 and denoted by ε. The canonical processes contributing to the isotopes are listed under the isotope mass. The patterns for Sr, for the normalization to ^{86,88}Sr, correspond to relative deficiencies in the *p*-process ^{84}Sr (or uniform excesses in ^{86,87,88}Sr). ^{87}Sr (triangle) has been corrected for decay of ^{87}Rb for 4.6 AE. [after PAPANASTASSIOU and WASSERBURG (1978) and PAPANASTASSIOU *et al.*, (1978)].

best estimates of the early solar system value using the $^{86}Sr/^{88}Sr$ normalization. This appears to favor the case for an ^{84}Sr deficiency, however it cannot be considered a strong argument. Alternate interpretations consistent with the Sr data include (a) a deficiency in the *r*-process component in ^{88}Sr, or (b) excesses in ^{86}Sr and in ^{87}Sr which would not coincide with those excesses expected from the canonical *s*-process. If we arbitrarily attribute the observed effects to a shortage in ^{84}Sr, this would require the addition of at least $\sim 2 \times 10^{-5}$ of the total Sr nuclei as exotic ^{84}Sr to the nebula to make up the average normal Sr composition. A shortage attributed to ^{88}Sr would correspond to a factor of 150 larger contribution of exotic Sr nuclei.

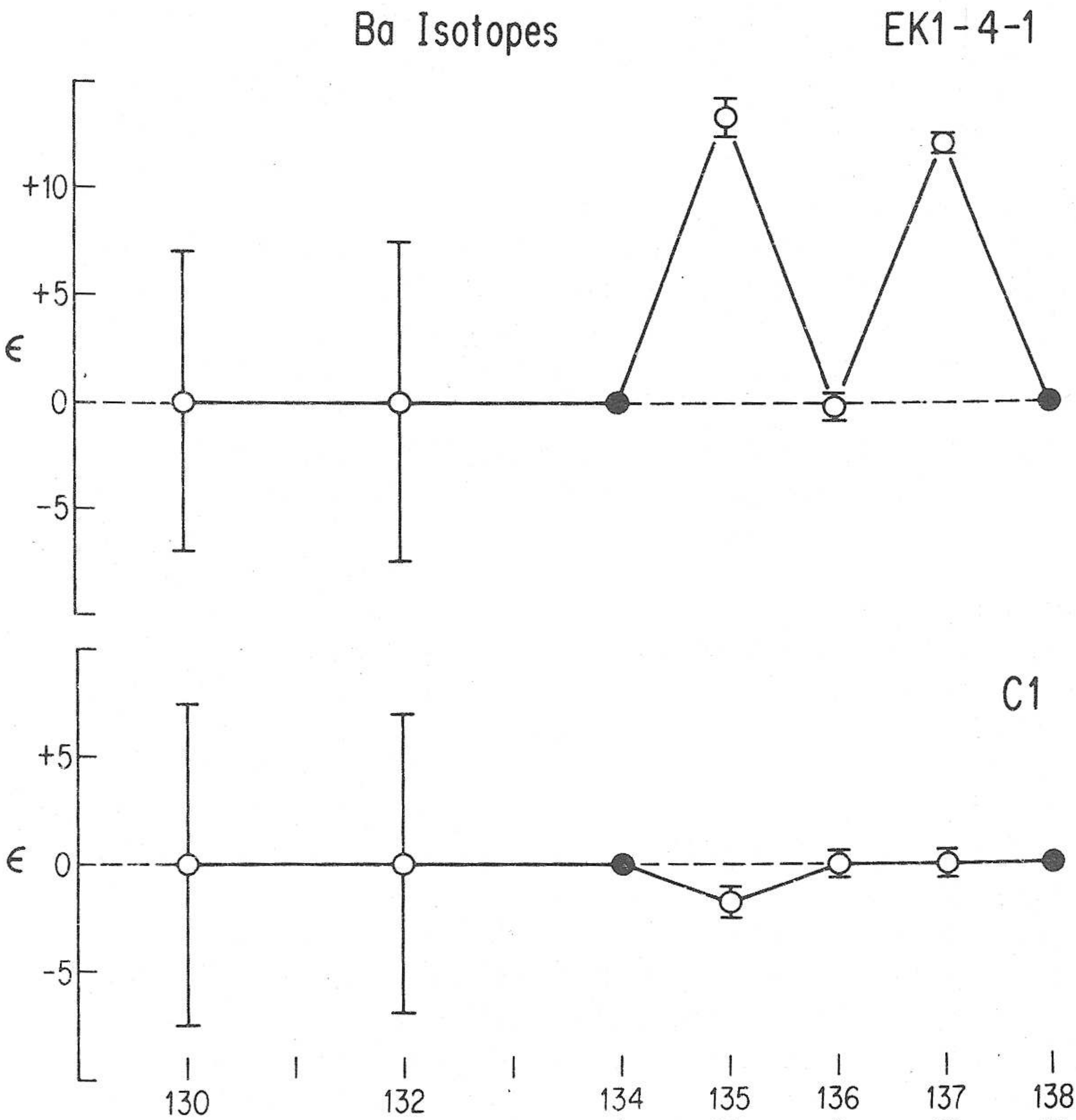

Fig. 14. Ba isotopic composition for the two FUN inclusions (*cf.* Figure 13 for conventions). EK-1-4-1 shows excesses in ^{135}Ba and ^{137}Ba while C-1 shows a small deficiency in ^{135}Ba. The less abundant *p*-process isotopes $^{130, 132}Ba$ show no effects but with significantly worse precision. C-1 shows no well-defined deficiencies in ^{137}Ba mirroring the excesses in EK-1-4-1. [after McCULLOCH and WASSERBURG (1978a)].

Barium. The data for Ba in C-1 and EK-1-4-1 (McCULLOCH and WASSERBURG, 1978a) are shown in Figure 14, normalized to ^{134}Ba and ^{138}Ba. Sample C-1 appears to be normal for all isotopes within errors except for a small but significant deficit of $\varepsilon 135 = -1.8$ at ^{135}Ba. Sample EK-1-4-1 shows a very different pattern with marked excesses $\varepsilon 135$ and $\varepsilon 137$ of 13 and 12 (parts in 10^4) respectively. All other isotopes for these samples have terrestrial abundances. Comparison of the data for these objects indicates that EK-1-4-1 has some nuclei added to the terrestrial value at masses 135 and 137 while C-1 appears to be the same as terrestrial except for a deficiency at mass 135. Inspection of the chart of the nuclides in the region of Ba (see Figure 15) shows that the isotopes $^{130, 132}$Ba are attributed to the *« p »* process. Neither of the FUN inclusions shows discernible effects at these masses within the larger uncertainties due to their low abundances. The other shielded isotopes $^{134, 136}$Ba are pure *« s »* process and $^{135, 137, 138}$Ba are dominantly *« s »* process, but ^{135}Ba and ^{137}Ba have significant *« r »* process components. The pattern exhibited by Ba in EK-1-4-1 strongly indicates the addition of *« r »* process material with a ratio $(^{137}\text{Ba}/^{135}\text{Ba})_r \sim 1.6$. This value is close to the observed terrestrial value. This should be compared with average *« r »* process production estimates (*cf.* WARD, NEWMAN and CLAYTON, 1976). The addition of a comparable number of nuclei to ^{138}Ba as appear to be added at 135 and 137 would not cause a measurable effect since ^{138}Ba is ~ 10 times more abundant.

The general discussion of the significance of deficiencies has been presented earlier. The simplest explanation is that the solar system Ba had an original isotopic composition which was deficient in ^{135}Ba and ^{137}Ba and that these isotopes were then added in fixed relative proportions to produce the average solar value and other mixtures with both residual deficiencies or excesses. In this case we would expect the observed deviations to follow the rule $\varepsilon 135 = (^{137}\text{Ba}/^{135}\text{Ba})_\odot \, (^{135}\text{Ba}^*/^{137}\text{Ba}^*) \, \varepsilon 137$. From the EK-1-4-1 data and terrestrial values we would expect for C-1 deficiencies in 137 about equal to 135 within ~ 10 %. The Ba spectrum of C-1 shows a deficiency in 135 but does not appear to include deficiencies at both 135 and 137 which are complementary to the excesses at these mass numbers as observed in EK-1-4-1.

It is possible to attribute a deficiency to ^{137}Ba in C-1 just within the limit of errors and with the same $(^{137}\text{Ba}^*/^{135}\text{Ba}^*)$ ratio for the injected matter as for EK-1-4-1. A possible explanation is that the ^{135}Ba deficiency in C-1 might be due to a holdup at ^{135}Cs ($\tau_{1/2} = 2.3 \times 10^6$ yr). The presence of ^{135}Cs would be compatible with the time scale for late injection defined by ^{26}Al. The ^{135}Ba deficiency is of great importance as it provided the first lower limit for the fraction of intermediate atomic weight nuclei which had to be added to the presolar system material to bring it up to the average value. The amount of ^{135}Ba required is $\sim 0.5 \times 10^{-4}$ atoms relative to a cosmic abundance of $\text{Si} \equiv 10^6$. This is commensurate in magnitude with the observed abundance of ^{129}I which is $\sim 10^{-4}$ atoms on the same scale (see Figure 15).

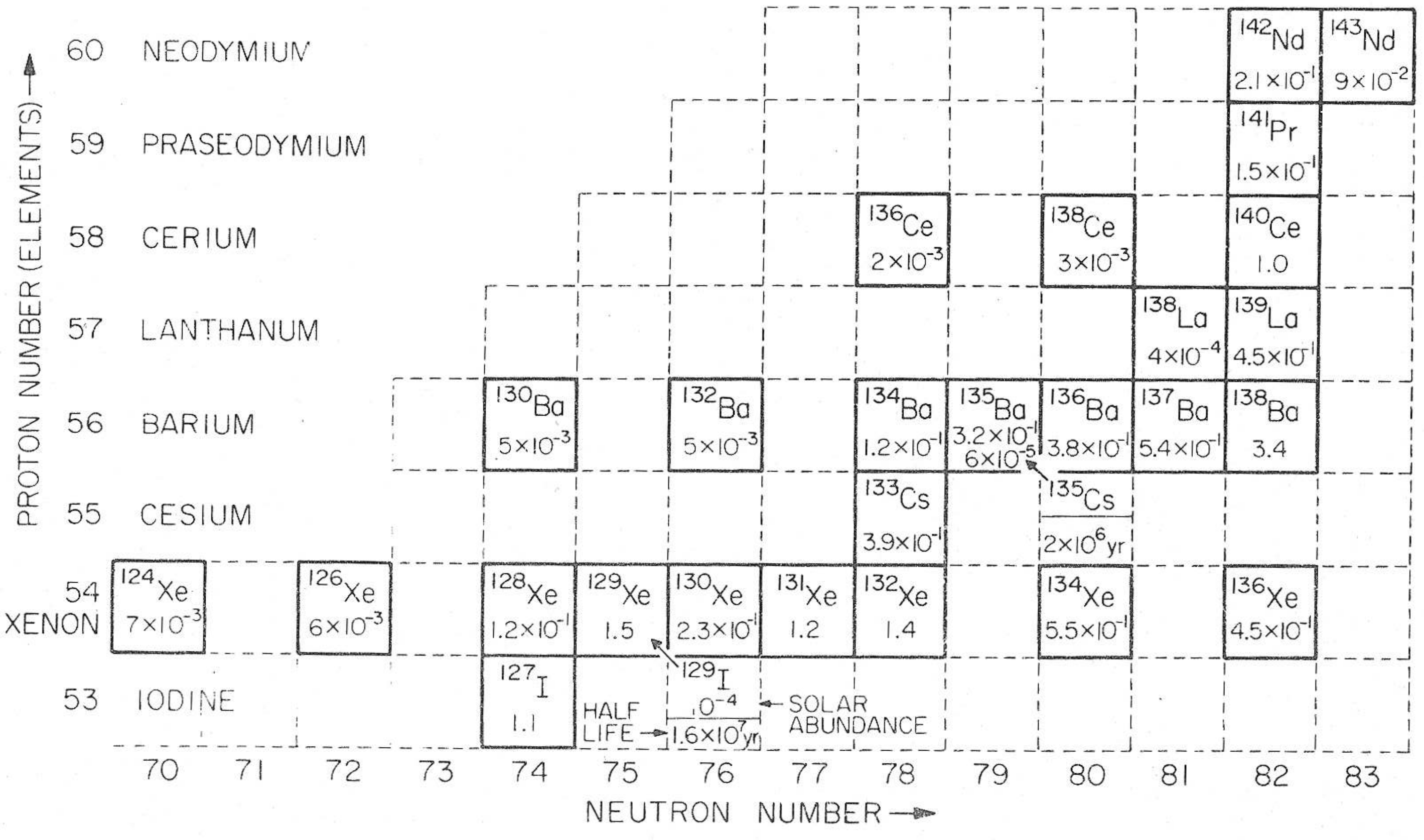

Fig. 15. Chart of the nuclides in the vicinity of I and Ba (*cf.* Figure 11 for conventions). Note the low abundance of *p*-process isotopes ^{130,132}Ba, the shielded isotopes ^{134,136}Ba, and contribution of 6.5×10^{-5} at ^{135}Ba. This appears to represent the minimum amount of ^{135}Ba to the solar nebula in the last event, and is comparable to the abundance of ^{129}I. The possibility of ^{135}Cs decay is hinted at from the data on inclusion C-1.

Neodymium. In general, isotopic anomalies in the rare earth elements are of special importance because of the fact that these elements are not readily subject to chemical fractionation. As a result, the number of exotic nuclei in most of the rare earth elements should be a reliable measure of the composition of injected material. The data for Nd (McCULLOCH and WASSERBURG, 1978a) in samples C-1 and EK-1-4-1 normalized to ^{142}Nd and ^{144}Nd are shown in Figure 16. It can be seen that the Nd in C-1 has normal composition to within analytical errors. In contrast, the results

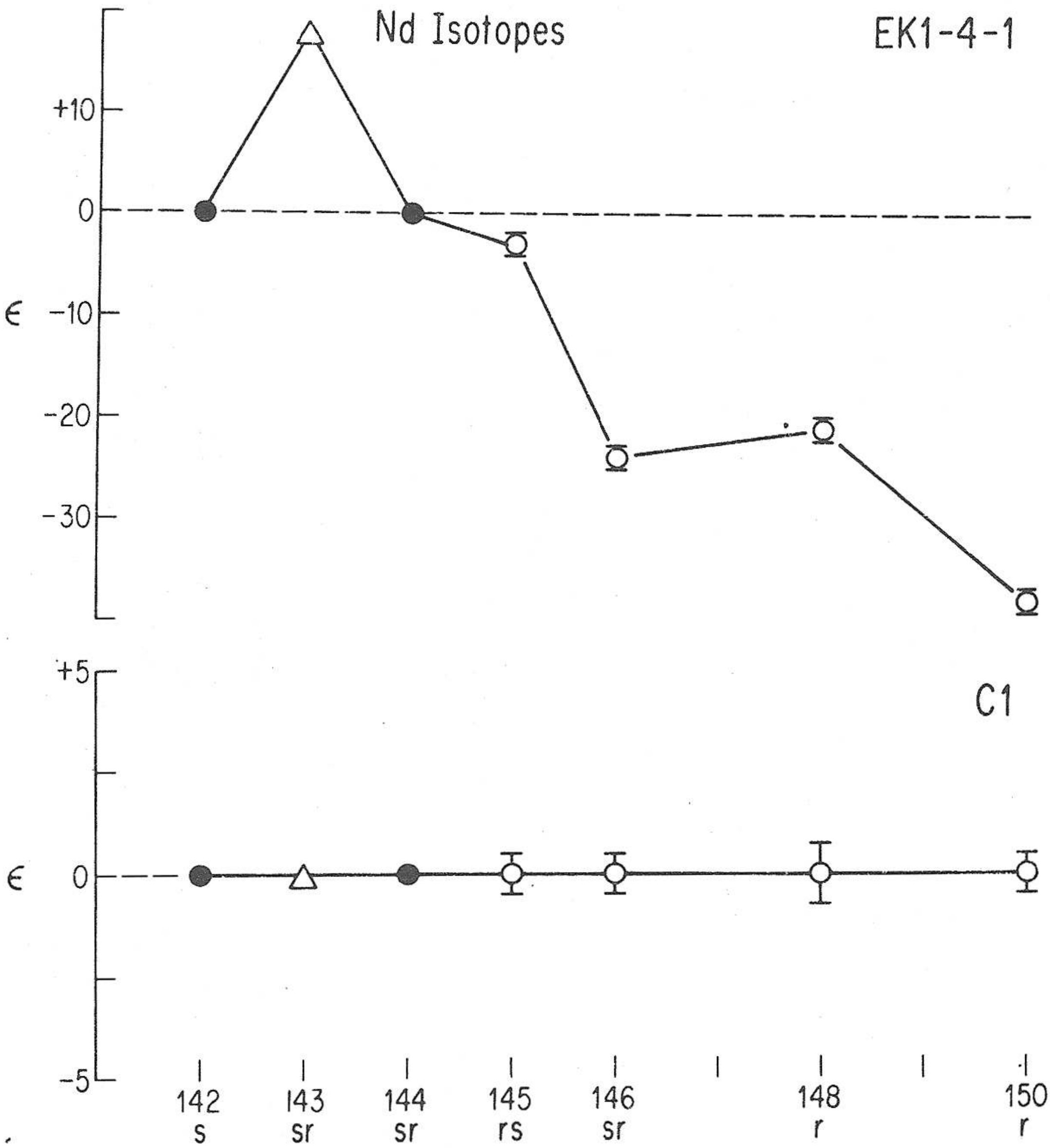

Fig. 16. Nd isotopic composition in FUN inclusions using a $^{142, 144}$Nd normalization (*cf.* Figure 13 for conventions). The pattern for Nd shows no regularities corresponding to the *s* and *r* processes indicated below mass numbers and indicates the need for alternate normalizations (*cf.* Figure 18). Nd composition for C-1 is normal. Note the absence of *p*-process isotopes in Nd. Data for ^{143}Nd (triangle) have been corrected for ^{147}Sm decay. [after McCULLOCH and WASSERBURG (1978a)].

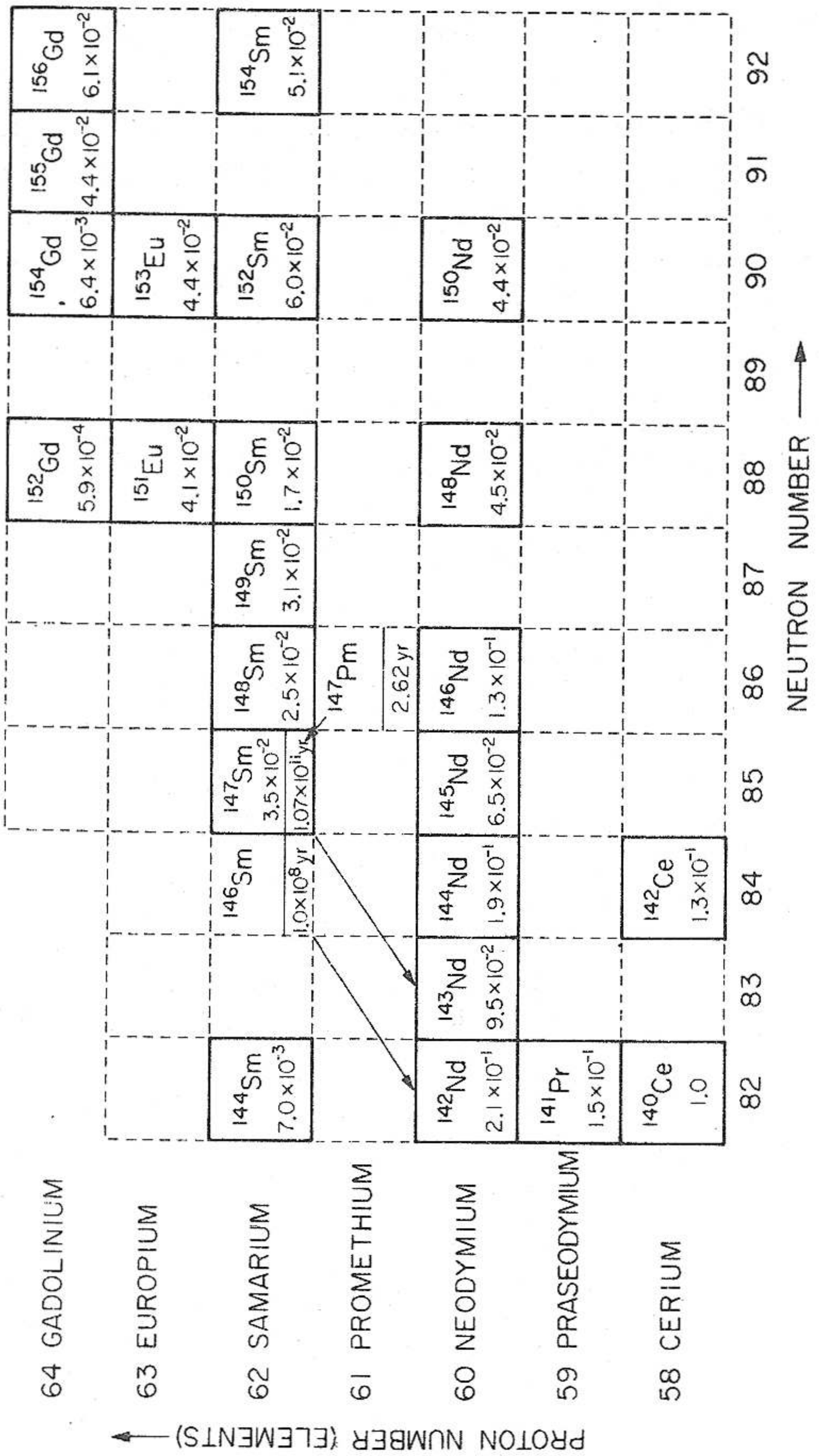

Figure 17. Chart of the nuclides for Nd and Sm (*cf.* Figure 11 for conventions). Note that for Nd only ^{142}Nd is shielded from the *r*-process, and that no *p*-process Nd isotopes exist. For Sm, ^{144}Sm is a relatively abundant *p*-process isotope while ^{148}Sm and ^{150}Sm are shielded by their Nd isobars.

for EK-1-4-1 show large deviations from the normal values. The magnitudes of the effects are very large for several isotopes. However, in this representation there are no regularities which would indicate simple addition of exotic nuclei to normal Nd. As discussed in the introduction, the choice of two isotopes for normalization is arbitrary and a selection which is more readily interpretable from nuclear systematics is preferable. From the data on Ba in EK-1-4-1 there is strong indication of addition to the unshielded isotopes for this sample. Inspection of the chart of the nuclides in the region of Nd (see Figure 17) shows that only ^{142}Nd is shielded (by ^{142}Ce). If we assume that addition to all the unshielded isotopes also occurs for Nd, we may obtain a revised form of the deviations with an additional assumption of the magnitude of the yield at another isotope. Taking a contribution to ^{144}Nd of $\varepsilon 144 = 18$ (18 parts in 10^4), we obtain the relative spectrum with additions in all the unshielded isotopes as shown in Figure 18. This representation is in full agreement with the model of «*r*» process excess (or *s* deficiency) inferred from the Ba results on the same sample.

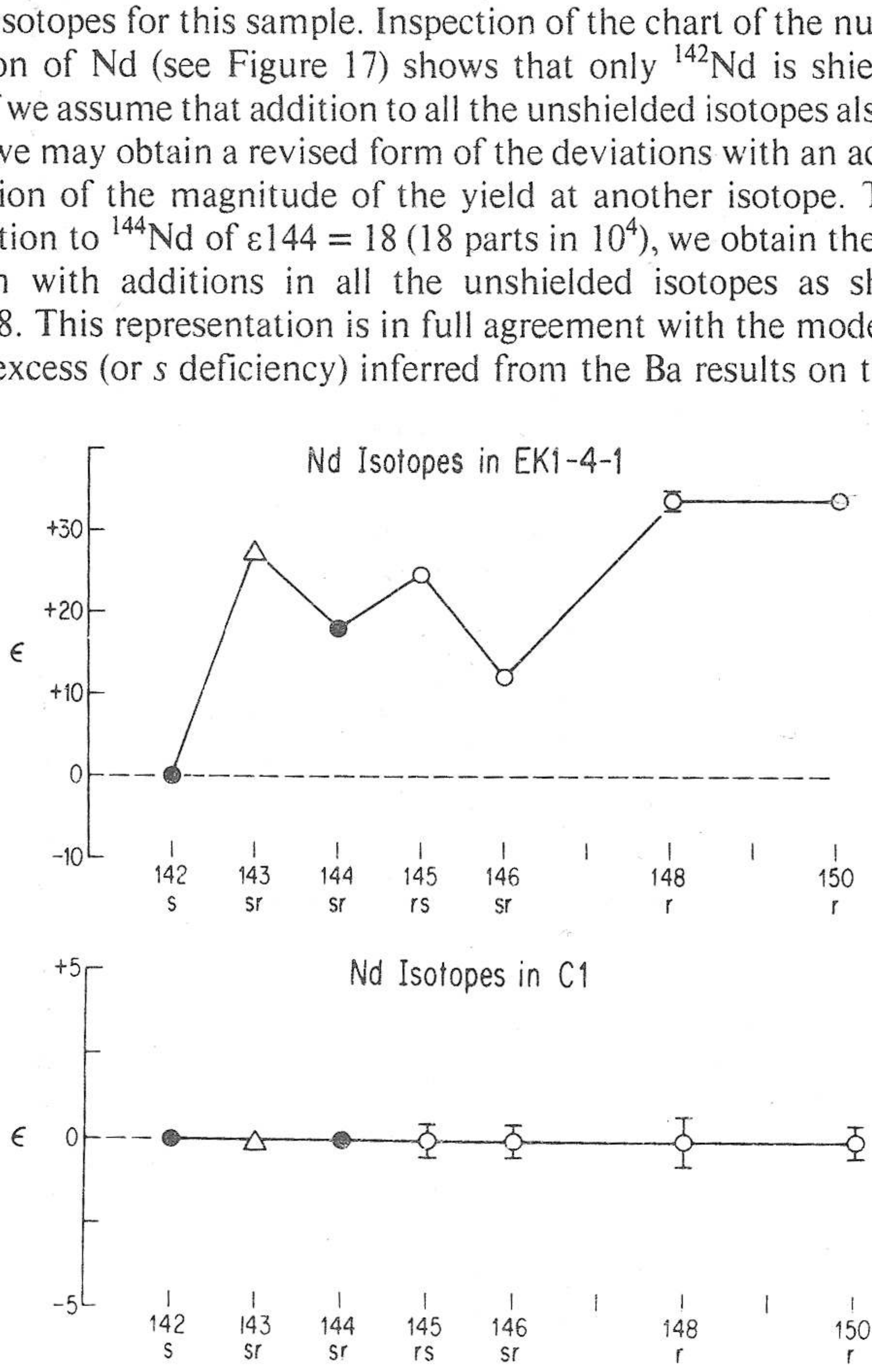

Fig. 18. Nd isotope composition in FUN samples using ^{142}Nd and ^{144}Nd for normalization after adding 18 parts in 10^4 to ^{144}Nd for EK-1-4-1 to reflect expected *r*-process contributions to this isotope also. Resulting pattern clearly reflects roughly commensurate relative excesses in the unshielded Nd isotopes (*cf.* Figure 16). Inclusion C-1 retains its normal composition (no assumption of ^{144}Nd addition was made). [after McCULLOCH and WASSERBURG (1978a)].

The absence of any effect in Nd from C-1 is somewhat surprising, however it should be noted that the deficiency in ^{135}Ba is quite small in magnitude. To explain both the C-1 and EK-1-4-1 data by addition of exotic material to a batch of «standard» material which is slightly deficient in some isotopes, and assuming no chemical fractionation between Ba and the rare earth elements (REE), would require that the contribution of exotic nuclei at Nd ($Z = 60$) is somewhat less than at Ba ($Z = 56$).

Samarium. Observations on Sm are particularly important as this ele-

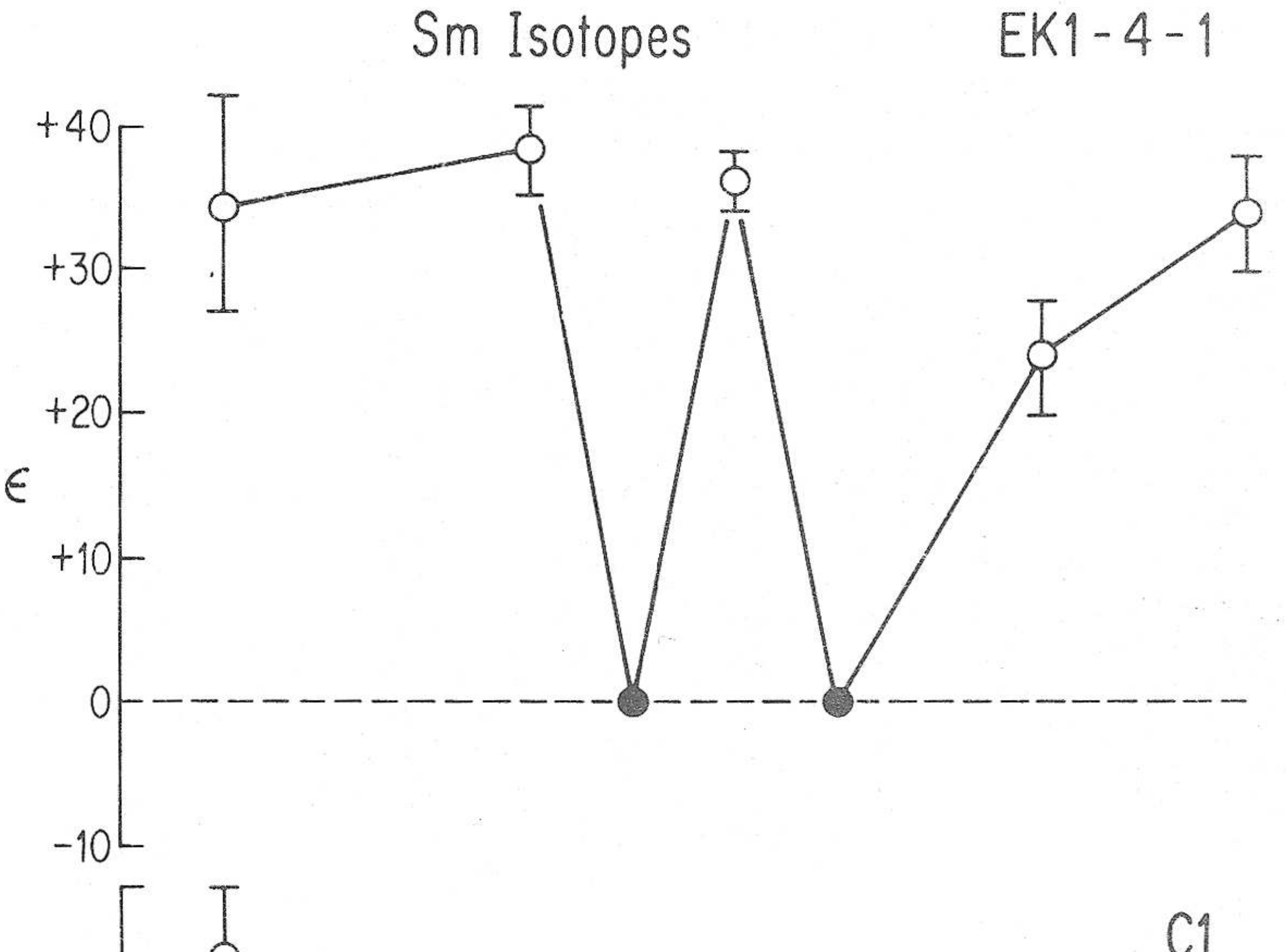

Fig. 19. Sm isotopic composition in the two FUN inclusions normalized to the two *s*-process only isotopes ^{148}Sm and ^{150}Sm (*cf.* Figure 11 for conventions). Sample EK-1-4-1 shows excesses of relatively equal magnitude in the unshielded isotopes and in the *p*-process isotope 144. Sample C-1 shows only an excess in the *p*-process isotope, indicating that *r* and *p* process effects can be decoupled. [C-1 and EK-1-4-1 data after McCULLOCH and WASSERBURG (1978b) and EK-1-4-1 data after LUGMAIR *et al.*, (1978)].

ment has three shielded isotopes, one of which (^{144}Sm) is attributed to the «*p*» process and the other two (^{148}Sm, ^{150}Sm) are pure «*s*» process. Sm is the only rare earth element with two pure «*s*» isotopes and can thus be used to test models of «*r*» process addition or enhancement relative to «*s*» process nuclei as first indicated by the observations on Ba. The chart of the nuclides in the neighborhood of Sm is shown in Figure 17. The Sm isotopic composition for the samples C-1 and EK-1-4-1 are shown in Figure 19 normalized to 144 and 148. This choice of normalization is the preferred one and most clearly exhibits the nature of the anomalies (McCULLOCH and

TABLE 1

Nucleosynthetic Components in Ba, Nd and Sm for EK-1-4-1

Nuclide	$N(A)$[a]	$\sigma(A)$[b]	$N_S(A)$[c]	$N_r(A)$[d]	$\varepsilon(A)$[e]	$\varepsilon(A)x$[f] / $N(A)10^{-4}$	$\frac{\varepsilon(A)N(A)}{\varepsilon(154)N_r(A)}$[g]
^{130}Ba	0.005[m]	2000[k]	0.0	0.0	0.0	0.0	—
^{132}Ba	0.005[m]	650[k]	0.0	0.0	0.0	0.0	—
^{134}Ba	0.116	225[h]	0.116	0.0	0.0	0.0	—
^{135}Ba	0.316	470[i]	0.055	0.261	13.4	4.23×10^{-4}	0.47
^{136}Ba	0.375	70[h]	0.375	0.0	0.0	0.0	—
^{137}Ba	0.543	72.6[i]	0.363	0.180	12.3	6.68×10^{-4}	1.08
^{138}Ba	3.44	5.7[h]	3.30	0.14	0.0	0.0	—
^{142}Nd	0.211	45[j]	0.211	0.0	0.0	0.0	—
^{143}Nd	0.095	332[i]	0.025	0.07	27.8[l]	2.64×10^{-4}	1.1
^{144}Nd	0.186	67[i]	0.116	0.07	18.0	3.35×10^{-4}	1.39
^{145}Nd	0.065	485[i]	0.016	0.049	24.9	1.61×10^{-4}	0.96
^{146}Nd	0.134	105[i]	0.069	0.065	12.0	1.61×10^{-4}	0.72
^{148}Nd	0.045	210[k]	0.0	0.045	33.6	1.50×10^{-4}	0.98
^{150}Nd	0.044	240[k]	0.0	0.044	33.6	1.47×10^{-4}	0.98
^{144}Sm	0.007[m]	120[k]	0.0	0.0	34.4	0.24×10^{-4}	—
^{147}Sm	0.035	1150[k]	0.006	0.029	38.5	1.34×10^{-4}	1.35
^{148}Sm	0.025	260[k]	0.025	0.0	0.0	0.0	—
^{149}Sm	0.031	1620[k]	0.004	0.027	36.5	1.14×10^{-4}	1.23
^{150}Sm	0.017	370[k]	0.017	0.0	0.0	0.0	—
^{152}Sm	0.060	450[k]	0.014	0.046	24.1	1.46×10^{-4}	0.92
^{154}Sm	0.051	380[k]	0.0	0.051	34.3	1.76×10^{-4}	1.0

[a]Cosmic abundance (per Si = 10^6), CAMERON (1973a). [b]30 KeV cross sections (millibarns). [c]*s*-process abundance. [d]*r*-process abundance given by $N_r(A) = N(A) - N_s(A)$. [e]Deviations from normal, in parts in 10^4, from McCULLOCH and WASSERBURG (1978a, b). [f]Excess atoms. [g]Ratio of excess atoms to cosmic *r*-process atoms, normalized to ^{154}Sm. [h]MUSGROVE, BOLDMAN and MACKLIN (1976). [i]HOLMES, WOOSLEY, FOWLER and ZIMMERMAN (1976). [j]CONRAD (1976). [k]ALLEN, GIBBONS and MACKLIN (1971). [l]Corrected for ^{147}Sm decay for an age of 4.56×10^9 years. [m]*p*-process only nuclide. [after McCULLOCH and WASSERBURG (1978b)]

WASSERBURG, 1978b). Data for Sm on EK-1-4-1 have also been reported by LUGMAIR, MARTI and SCHEININ (1978); these workers have also confirmed the Nd measurements on EK-1-4-1 of McCULLOCH and WASSERBURG (1978a). The data for C-1 indicate that all Sm isotopes are terrestrial in composition with the exception of the «*p*» process ^{144}Sm, which has an enrichment of $\varepsilon 144 = 15 \pm 5$. In contrast EK-1-4-1 shows large excesses for the four unshielded isotopes and also an excess for the «*p*» process ^{144}Sm. These results on Sm provide additional strong evidence for «*r*» process enrichment relative to the «*s*» isotopes.

Summary of Mg, Si, Ca, Sr, Ba, Nd, and Sm in two FUN samples. The isotopic analyses for Ba, Nd, and Sm on C-1 and EK-1-4-1 which we have discussed here were performed on bulk samples of these Ca-Al rich inclusions and on separated mineral phases. Each of the samples analyzed had different ratios of Ba/Nd/Sm. The different phases within each inclusion showed the same peculiar isotopic composition for these elements. The isotopic composition of the two inclusions was distinct. This observation is in support of similar results for Mg, Ca, and Sr and distinct from that observed for oxygen. We, therefore, conclude that each inclusion represents a sample of isotopically peculiar material which was made isotopically homogeneous for refractory elements at the time of crystallization. Presolar dust grains, which may have supplied material making up these inclusions, have been destroyed as distinguishable objects for the major oxide phases.

The elements Ba, Nd, and Sm in EK-1-4-1 have isotope shifts which can all be explained by the addition of a generic «*r*» type component to normal material or the enhancement of «*r*» process relative to «*s*» process material. The relative abundances of the added nucleosynthetic component is rather close to that for average «*r*» process nuclei (see Table 1). The Sm data, however, also require a concomitant enhancement in the «*p*» process nuclide in Sm which is not observed in Ba. Inspection of Figures 15 and 17 and Table 1 shows that the solar abundances of ^{144}Sm, ^{130}Ba, and ^{132}Ba are comparable so the addition of the same number of $^{130, 132}$Ba* nuclei as ^{144}Sm* nuclei to solar material should produce similar effects. While the uncertainties in these rare Ba isotopes are relatively large, there is no hint of any effect. It is not clear why ^{144}Sm should be preferentially enhanced considering the abundances of possible progenitors for the «*p*» isotopes of Ba and Sm, whether by successive *p*, γ or γ, *n* reactions. This may suggest that the late «*p*» type process was very specific or it may reflect the fact that ^{144}Sm is a very abundant Sm isotope and hence is always favored. More detailed theoretical analysis of the «*p*» process is clearly needed. An investigation of «*p*» process addition to heavier rare earth elements in C-1 and EK-1-4-1 should aid in clarifying this problem of process yields. The presence of ^{144}Sm excesses in C-1, which is otherwise normal and in EK-1-4-1, which also has excesses in the unshielded isotopes, indicates that the «*p*» and «*r*» components are not coupled but are

independent. This rule, if valid, should place strong constraints on possible astrophysical sites and processes for «*p*» and «*r*» nuclide production.

The isotopic patterns for Ba, Nd, and Sm appear to be most readily recognizable ones. Insofar as the patterns are dominated by excesses of unshielded isotopes relative to shielded isotopes, it is reasonable to associate them with processes with very high neutron fluxes. This may be some type of «*r*» process. It is difficult to conceive of proton bombardment mechanisms which would yield these effects unless there were some special mechanism for abundant neutron production. In consideration of all the isotopic anomalies reported here, the results on the heavy elements seem to point strongly toward a supernova.

Some of the results could be explained by the spontaneous or induced fission of an unknown progenitor. As the isotopic effects are independent of the relative chemical abundances within the individual phases in both inclusions, the fission would have had to take place prior to crystallization. Using the deficiency of ^{135}Ba in C-1 as an estimate, this would require an abundance of $\sim 3 \times 10^{-5}$ fissionable parent nuclei (100 % fission) in units of 10^6 Si atoms. The fission hypothesis does not explain the ^{144}Sm excess and could not be the source of the anomalies found at low *Z*.

In considering the very short time scale as determined both by ^{26}Al and ^{107}Pd, there must have been late production and injection of nuclei with a wide range in mass. It is likely that some very heavy fissionable nuclei (uranium and transuranics) were produced which contributed to the material added to the proto-solar cloud. Such nuclei could alter the isotopic effects to be expected from a «simple *r* process» addition, and should be taken into account in any detailed theory. However, if the yield curve for the injected nuclei is in any way similar to the solar abundances, we would expect such contributions to be comparatively small.

We note a clear distinction in interpretation if we extend our considerations to the observation on the lower atomic number elements Mg, Si, Ca, and Sr for the two FUN inclusions. For example while there is a clear indication of *p* excess, *s* deficiency, and *r* excess for Sm in EK-1-4-1, the Sr shows a *p* deficiency or *s* excess or *r* deficiency. The Ca effects are not attributable to any clearly defined process. The Mg shows a deficiency in ^{24}Mg, an excess in ^{25}Mg, or a deficiency in ^{26}Mg. Again there is no clearly defined process from purely nuclear considerations. This apparent lack of correlation between isotopes of different elements in the same samples, both in the types of processes involved as well as in the sign of the effects, indicates that a mixture of specific nucleosynthetic processes, astrophysical production sites, and models of extraction of materials from a star are required to interpret the effects. Alternatively the isotopic effects depend in detail on the specific synthesis processes operating possibly in a restricted fashion or on pre-existing seed nuclei, so that the effects cannot be directly associated with the canonical, average *r*, *s*, and *p* processes. For some nuclei there will have to be a better definition of the production scheme.

There is a further distinction in the presence of isotopic fractionation for some of the low Z elements (O, Mg, Ca, and Si) which is not apparent in the heavier elements. This is undoubtedly a reflection of the much greater atomic weight but may also depend on the complex nature of the chemical (and ion) kinetics and the distribution of these elements in the local parcels of gas and dust from which the inclusions formed. So far it has not been possible to predict which elements in the FUN inclusions should show marked fractionation. The basic reason for the presence of nuclear anomalies in matter which shows strong fractionation in some elements is not obvious.

TIME SCALES

One of the basic problems concerning the observed isotopic heterogeneities is the time to be associated with the production of the various nuclides relative to the formation of the solar system. The identification of extinct radioactive nuclides in early solar system material is the principal means of determining the times of production. It is plausible that the time scale for chemical and isotopic mixing between gas and dust is very long under cool interstellar cloud conditions. In addition, dust grains and possibly macroscopic fragments could have been formed in different stellar sites and preserved in the aggregation of larger bodies. These considerations may complicate interpretation of some observations. If it can be shown that relatively short-lived nuclides were present in macroscopic objects which formed as isotopically homogeneous bodies within the solar system, then it is possible to establish a time scale that links the time of nucleosynthetic processes with the solar system time scale. As emphasized by D.D. Clayton (*cf.* CLAYTON, 1975), it is possible that a short-lived nuclide decayed in an object, say a dust grain, prior to being incorporated into the solar system. In this case only a lower limit to the time scale can be obtained and there is no direct connection between a nucleosynthetic event and the solar system chronology.

Until recently the only well identified short-lived nuclides were extinct ^{129}I ($\tau_{1/2} = 16 \times 10^6$y) and the not quite extinct ^{244}Pu ($\tau_{1/2} = 83 \times 10^6$ y). The evidence for the existence of ^{129}I is the presence of excesses of ^{129}Xe relative to other Xe isotopes and the correlation of this excess with the presence of ^{127}I (*cf.* REYNOLDS, 1960, 1963; JEFFERY and REYNOLDS, 1961). The evidence for ^{244}Pu is from observations of the unshielded neutron rich isotopes of Xe with a unique abundance pattern for ^{131}Xe, ^{132}Xe, ^{134}Xe, ^{136}Xe (*cf.* ROWE and KURODA, 1965). This unique Xe spectrum is correlated with the presence of fission tracks in minerals rich in U, Th, and REE (WASSERBURG, HUNEKE and BURNETT, 1969; LEWIS, 1975). Plutonium is known to have chemical characteristics similar to Nd and Sm and should thus be associated with these elements. The Xe spectrum of ^{244}Pu spontaneous fission was found to correspond precisely with the unique Xe spectrum found earlier in some meteorites (*cf.* ALEXANDER, LEWIS, REYNOLDS and MICHEL,

1971). Some of the meteorites which contain the ^{224}Pu SF products are obviously the result of crystallization from a silicate melt and the product of planetary differentiation (*cf.* Angra dos Reis, HOHENBERG, 1970: WASSERBURG, TERA, PAPANASTASSIOU and HUNEKE, 1977). There must of course be some ^{244}Pu remaining today and a hint exists in one preliminary experiment.

The existence of ^{244}Pu in the early solar system and in planetary objects does not by itself provide a strict time scale because of the long half life. The estimated abundance of ^{244}Pu relative to ^{232}Th at the time of formation of the solar system is $^{244}Pu/^{232}Th \sim 5 \times 10^{-3}$ (PODOSEK, 1970). This value in conjunction with a plausible relative production rate for these two nuclides yields a time scale of $\sim 3\text{-}4 \times 10^8$ y. ^{129}Xe excesses have been found in a variety of meteorites including basaltic achondrites and in stony and sulphide inclusions in iron meteorites which are planetary differentiates (PODOSEK, 1972; WASSERBURG *et al.*, 1977; NIEMEYER, 1978). This ^{129}Xe is strongly correlated with ^{127}I and it must therefore be considered as resulting from the *in situ* decay of ^{129}I in the solar system. The ^{129}I must have been included not only in aggregates of more primitive material (including Allende inclusions) but in differentiated planetary objects (iron tmeteorites and a eucrite) after their formation. The abundance of ^{129}I relative to ^{127}I at the time of solar system formation is $^{129}I/^{127}I \sim 1 \times 10^{-4}$. This implies a time interval of $1\text{-}2 \times 10^8$ y between the last period of ^{129}I production. Until very recently the $^{129}I/^{127}I$ and $^{244}Pu/^{232}Th$ abundances were used in conjunction with the $^{235}U/^{238}U$ and $^{238}U/^{232}Th$ abundances to construct self consistent time dependent models of galactic nucleosynthesis (*cf.* SCHRAMM and WASSERBURG, 1970). The rate of «*r*» type nucleosynthesis with time as determined by this approach depends on the $^{244}Pu/^{232}Th$ ratio. In particular the question of whether there was a terminal spike is sensitive to the $^{244}Pu/^{232}Th$, $^{235}U/^{238}U$, and the $^{129}I/^{127}I$ ratios. However, in all cases the last time of production is governed by the $^{129}I/^{127}I$ ratio since ^{129}I has the shortest half life of these nuclides.

For many years there have been intermittent searches for evidence of shorter lived nuclides. Searches were carried out for ^{26}Al ($\tau_{1/2} = 0.7 \times 10^6$y) with the daughter product ^{26}Mg and ^{107}Pd ($\tau_{1/2} = 6.5 \times 10^6$ y) with the daughter product ^{107}Ag. Until recently no positive results had been brought forward which withstood careful tests of verification. If the observed ratio of $^{129}I/^{127}I \sim 10^{-4}$ is the result of sudden production (with a ratio of production rates of order unity) followed by free decay without any further contributing nucleosynthetic events, then a nuclide with a half life only half of that of ^{129}I would only have an abundance relative to a stable isotopic cogener of 10^{-8}. Such a disparity in half lives is so enormous that a free decay interval of $\sim 10^8$ y would absolutely prohibit the persistence of shorter lived nuclides. It should be noted that in the case of ^{26}Al, the solar value is $(^{27}Al/^{26}Mg)_\odot \sim 0.7$. If $^{26}Al/^{27}Al$ is as high as 10^{-3} then, for the solar system, the total increase in ^{26}Mg due to ^{26}Al decay would be 0.07 %. This means that to observe any effects it is necessary to have the most ancient materials and they must also be highly enriched in Al relative to Mg.

For almost two decades the astrophysical and cosmochemical communities were quite satisfied with a scenario in which the interstellar cloud from which the solar system formed was isolated from either local or galactic nucleosynthesis for $\sim 1 \times 10^8$ y. Attempts by FOWLER, GREENSTEIN and HOYLE (1962) to account for D, Li, Be, and B by local bombardment did not appear acceptable both because of the observational data on isotopic homogeneity and the specialized geometry of the assumed bombardment by energetic solar protons. Some workers, particularly A.G.W. Cameron, had argued for a supernova trigger for the formation of the solar system (*cf.* CAMERON, 1963). The fall of the Allende meteorite in 1969 brought large quantities of material containing Ca and Al rich inclusions that approximated in chemical composition what had been calculated theoretically to be the early condensates from a hot medium of solar composition (LORD, 1965; GROSSMAN, 1972). Similar Ca-Al rich inclusions had first been recognized by CHRISTOPHE-MICHEL-LEVY (1968) in the Vigarano meteorite. The large mass of the Allende meteorite made the Ca-Al rich inclusions more readily available for scientific research. This meant that more « primitive » samples of early solar system material could be studied in contrast to the more evolved or more thoroughly mixed objects which had previously been available.

The existence of abundant samples, in conjunction with the development of more advanced methods used in the study of lunar samples, led to a new generation of high precision, high sensitivity meteorite studies. An extensive investigation by GRAY, PAPANASTASSIOU, and WASSERBURG (1973) showed that some of the Ca-Al rich inclusions in Allende were the oldest known objects in the solar system as manifested by the very low $^{87}Sr/^{86}Sr$ ratios. These workers indicated that a new search for ^{26}Al was necessary. CAMERON (1973b) proposed that this primitive character of Sr might not be due to a shorter time for ^{87}Rb decay in the solar system but rather incomplete mixing of « *s* » and « *r* » process material from a late injection. While this interpretation of the Sr results could not be substantiated, the possibility of a shortened time scale was again brought into consideration. This was greatly enhanced by the discovery by CLAYTON *et al.*, (1973) of isotopic anomalies in oxygen. A search of isotopic anomalies in Mg led to the discovery by GRAY and COMPSTON (1974) and LEE and PAPANASTASSIOU (1974) of distinct shifts in the isotopic abundance of Mg in some Allende inclusions which could not be simply explained by mass fractionation. Subsequent work by LEE, PAPANASTASSIOU and WASSERBURG (LPW and permutations; LPW, 1976, 1977; PLW, 1977; WLP, 1976, 1977) showed that in two inclusions there were isotopic anomalies in Mg which were due to excesses in ^{26}Mg ($^{26}Mg^*$) (relative to terrestrial Mg) and that these excesses were strongly correlated with the Al/Mg ratio of the coexisting mineral phases. Evidence of the existence of an extinct nuclide is provided by demonstrating that excesses of the daughter nuclide are present in several different objects when the other isotopes of the daughter element are unchanged. Demonstration that

the now extinct parent nuclide was present in an object at the time of its formation requires that the observed distribution of the daughter nuclide follows the chemical distribution of the parent nuclide in the different phases of that object (*cf.* Figure 20). This requires that objects be well preserved since the time of formation. Figure 21 shows the relationship of Mg isotopic composition and the Al/Mg ratio for coexisting phases formed at a single time from a piece of matter which was initially isotopically uniform. The time «zero» refers to the time at which the matter was isotopically uniform. If the isotopically uniform state (of say Mg and Al) refers to the whole solar nebula, then the lines correspond to times relative to «zero», and may therefore be used as a chromometer. As samples may be chemically fractionated at different times, the intercepts $(^{26}Mg/^{24}Mg)_I$ may differ for isochronous objects, but the slopes will be equal. If the solar nebula is heterogeneous (with regard to the isotopic composition of Mg and Al) then the demonstration of *in situ* decay is still possible but the decay scheme does not provide a chronometer for different samples of the solar nebula. However, it does provide an approximate chronometer for the time between production of the parent nuclide and the object if production rates and dilution factors can be estimated and if the half life is short. A striking example of this correlation was found in inclusion WA and is shown in

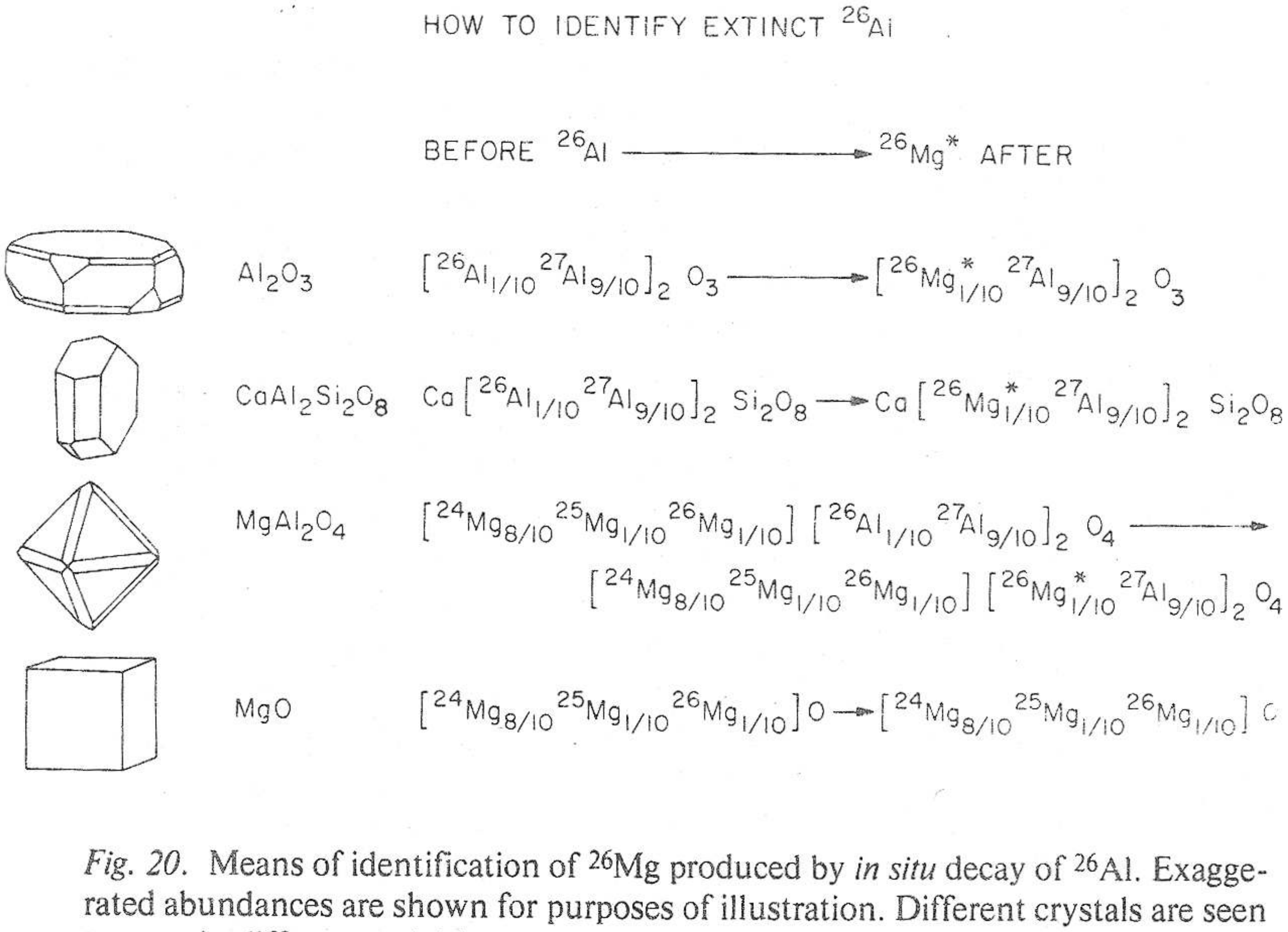

Fig. 20. Means of identification of ^{26}Mg produced by *in situ* decay of ^{26}Al. Exaggerated abundances are shown for purposes of illustration. Different crystals are seen to contain different stoichiometric ratios of Al/Mg but to have formed with a uniform Al isotopic composition consisting of 1/10 ^{26}Al and 9/10 ^{27}Al. The formulas are grouped so as to show that subsequent to decay the radiogenic $^{26}Mg^*$ replaces ^{26}Al atoms in the crystal lattices. If some of these mineral phases (and crystals) coexisted in one inclusion and were extracted, their $^{26}Mg/^{24}Mg$ ratio would correlate precisely with $^{27}Al/^{24}Mg$.

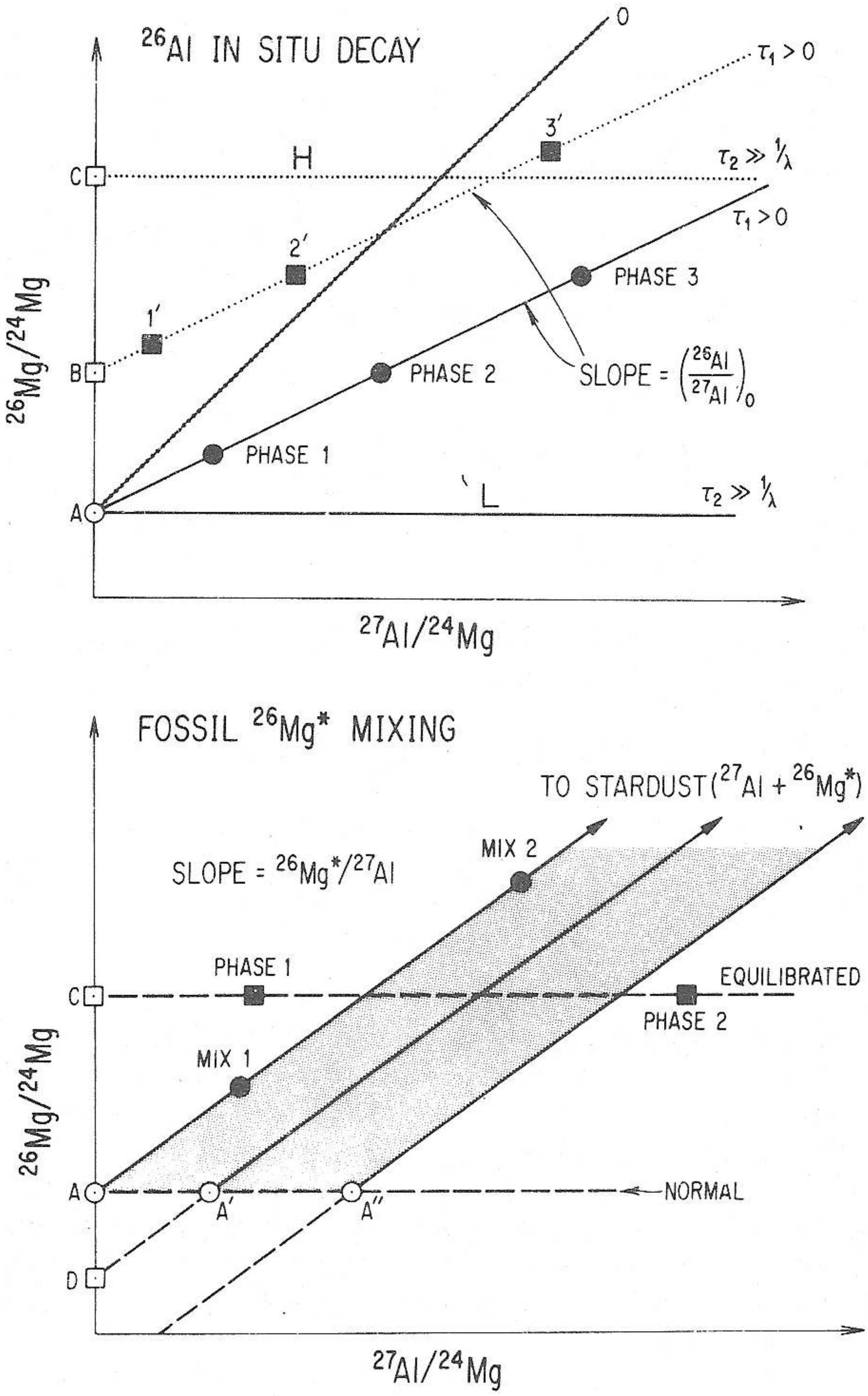

Fig. 21. (top) Al-Mg evolution diagram for two isochronous systems which were initially isotopically homogeneous. Phases in each system are colinear. Slopes correspond to $^{26}Al/^{27}Al$ in the systems. Differences in initial $^{26}Mg/^{24}Mg$ reflect differences in Al/Mg (high, low) in the parent reservoirs. (bottom) Systematics of mixing of normal material with stardust containing ^{27}Al and fossil $^{26}Mg^*$. Mixtures of stardust and of solar system materials containing Al and Mg will fall in the stippled area and will not in general define a straight line. [after LPW (1977)].

Figure 22. The plagioclase ($CaAl_2Si_2O_8$) has a high ratio of Al/Mg and yields an excess of ~ 8.5 % of ^{26}Mg relative to terrestrial Mg. This correlation was shown to hold for individual crystals (~ 100 μm) as well as for macroscopic samples of these crystals. By assuming that the ratio

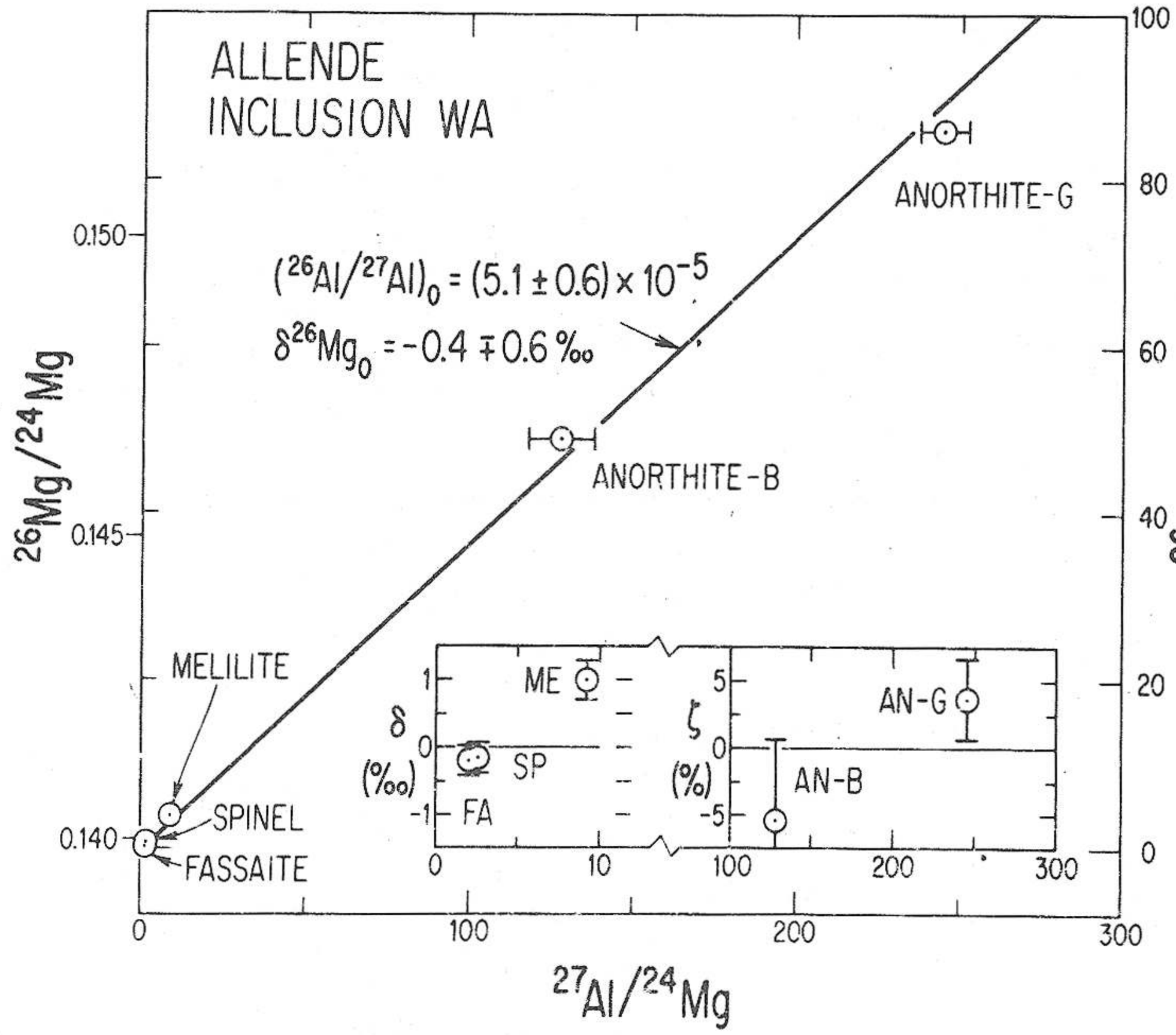

Fig. 22. ^{26}Mg-^{26}Al evolution diagram for coexisting mineral phases in Ca-Al-rich inclusion WA from Allende. Anorthite samples show very large excesses of ^{26}Mg (righthand axis) up to 85 permil correlating with $^{27}Al/^{24}Mg$. Melilite also shows significant ^{26}Mg excess. Insert shows deviations of the data from the best fit line; note that the melilite (ME) analysis is not on the best fit line. The slope of the line corresponds to an initial $^{26}Al/^{27}Al = 5 \times 10^{-5}$ at the time the inclusion formed. The initial Mg in the inclusion had normal $^{26}Mg/^{24}Mg$ composition. This correlation provides the basic evidence demonstrating the *in situ* decay of ^{26}Al in the early solar system. [after LPW, 1977].

$^{25}Mg/^{24}Mg$ was the same as terrestrial, it was possible to show that there was an apparent excess of ^{26}Mg in a phase (melilite) with an intermediate Al/Mg ratio in the same inclusion. The data point for the melilite lies close to, but is not on, the correlation line. This correspondence in $^{26}Mg^*$ with Al content was also found in an inclusion BG2-6 where the high Al phase is grossular ($Ca_3Al_2Si_3O_{12}$). This correlation has been substantiated in other Allende inclusions by independent workers using different techniques (BRADLEY, HUNEKE and WASSERBURG, 1978; HUTCHEON, STEELE, SMITH and CLAYTON, 1978; LORIN, SHIMIZU, CHRISTOPHE and ALLEGRE, 1977; LORIN and CHRISTOPHE, 1978). Some of the data are, however, of much lower precision so that strict tests of a linear correlation are not possible. In the case of inclusion WA, excesses of up

to 50 % in ^{26}Mg have been found (BRADLEY *et al.*, 1978). There are some clear violations of the correlation shown in Figure 22 (see PLW, 1976; and ESAT *et al.*, 1978). These are explainable by late stage melting of the inclusions, chemical alteration of the inclusions or incomplete mixing of ^{26}Al in the proto-solar nebula. Existence of ^{26}Mg excesses in meteorites other than Allende have been reported by LORIN *et al.*, (1977) and LORIN and CHRISTOPHE (1978) so that the evidence for ^{26}Al includes at least two meteorites. It appears that there is strong evidence for the existence of ^{26}Al in the early solar system at the time that centimeter-size objects were formed. There will have to be more extensive investigations to adequately document the case for *in situ* ^{26}Al decay (particularly for phases which intrinsically contain both Al and Mg in their crystal structure) and to obtain a better estimate of $^{26}Al/^{27}Al$ for the solar system. There is as yet no clear evidence for large $^{26}Al/^{27}Al$ ratios in samples which show general isotopic anomalies for many elements (such as C-1 and EK-1-4-1) and there is no correlation with the oxygen isotopic effects. Whether the ^{26}Al production is connected in time with the general isotopic anomalies (O, Mg, Ca, Kr, Sr, Xe, Ba, Nd, Sm) remains to be demonstrated. At present it appears that ^{26}Al was present in the solar system with an abundance of $(^{26}Al/^{27}Al) \sim 5 \times 10^{-5}$ in some objects but that ^{26}Al was not uniformly distributed. In some cases there has been serious alteration of the ^{26}Mg-^{26}Al parent-daughter relation by element redistribution due to chemical reactions between the early condensates and a later lower temperature gaseous regime.

The abundance ratio $(^{26}Al/^{27}Al)_{\odot}$ is an extremely sensitive indicator of the number of exotic nuclei added in a late stage. If the ratio $(^{26}Al/^{27}Al)_{SN}$ is the value in the source with due regard for dilution in the envelope, then this implies that at least a fraction $(^{26}Al/^{27}Al)_{\odot}/(^{26}Al/^{27}Al)_{SN}$ of the total Al in the solar system was injected by this late event. For an effective production ratio of $(^{26}Al/^{27}Al)_{SN} \sim 10^{-3}$ (ARNETT and WEFEL, 1978; TRURAN and CAMERON, 1978) and a solar system value of 5×10^{-5}, this corresponds to a contribution of at least 5 % of all the Al in the Sun.

The time scale determined by the existence of ^{26}Al in the solar system is at most 10×10^6 y and more reasonably $\sim 3 \times 10^6$ y. It follows that the time for the last injection of freshly synthesized nuclear material is at least an order of magnitude less than that obtained from ^{129}I. The time scale of 3×10^6 y is short compared to a galactic year and suggests the formation of the solar system in a region where many stars are being formed and which contains massive, rapidly evolving stars which can contribute fresh nuclear debris over the time the solar system is accumulating. An alternative possibility is that the early sun went through a phase of intense activity with a large flux (10^{23}p/cm^2) of energetic protons which produced ^{26}Al in a surrounding dust cloud (*cf.* LEE, 1978). It is not apparent how this mechanism could produce the observed excesses in unshielded isotopes of Ba, Nd, and Sm as discussed elsewhere in this paper.

The ^{26}Al time scale is extremely short; it is commensurate with the time scale for gravitational free fall and much shorter than the Kelvin-Helmholtz time. In order to understand the evolution from a state of highly dispersed material to collapse and the formation of the Sun and planets in a time of less than 10^7 years will demand imaginative and careful analysis. This condensation and formation time scale is required if the ^{26}Al is due to injection from external sources such as a supernova. The sequence of formation of the smaller planetary objects, the terrestrial and Jovian planets and the Sun is an open issue. There appears to be a major time difference between the formation of small objects (marbles to meteorite parent bodies) and the moon and the earth. If the young ages of the earth and moon ($\Delta T \sim 0.1$ AE) are not times of internal differentiation, then the terrestrial planets were accumulated from « meteoritic » debris long after the decay of ^{26}Al. This means that accumulation of silicate planetary bodies up to ~ 100 km was very rapid and the subsequent accumulation of larger terrestrial bodies was late, most plausibly after the formation of the sun and the Jovian planets. The absence of ^{26}Al in the earth, moon, Mars, Venus, and Mercury would again leave the initial heat sources in these bodies as a problem.

An abundance of $(^{26}\text{Al}/^{27}\text{Al}) \sim 5 \times 10^{-5}$ provides an intense heat source for planets which form promptly. This could explain the rapid melting of small asteroidal size objects (down to a few km) as well as for the larger terrestrial planets (*cf.* UREY, 1955). This abundance in the gas and dust of the solar nebula would also be a major source of ionizing radiation (CONSOLMAGNO, 1977). The applicability of ^{26}Al as a general planetary heat source and as a radiation source in the solar nebula must depend on the large scale abundance of this nuclide and the rate of planet formation. This can only be addressed by more extensive sampling of early solar system debris.

The issue regarding times scales is summarized in Figure 23. In the past few years we have gone from ~ 10^8 y to ~ 10^6 y for the last time of injection. It is clear that some of the intermediate-lived nuclei (*e.g.* ^{244}Pu) must come from somewhat earlier periods of nucleosynthesis ($1 - 4 \times 10^8$ y). Earlier contributions for ^{244}Pu are required in order to explain the solar $^{244}\text{Pu}/^{232}\text{Th}$ ratio since the production of ^{244}Pu (and ^{235}U) on a 10^6 y time scale is small, unless the production pattern for high mass ($A > 200$) nuclei is enriched by a factor of 100 relative to those at intermediate mass for the late stage addition. The longer-lived and stable nuclei must dominantly come from much more ancient events. The rate of nucleosynthesis as estimated from the most recent events which we sample in the solar system gives a clear hint of the granularity of the nucleosynthetic processes but does not yet permit a firm estimate of the rates of nucleosynthesis over the history of the galaxy. It is possible that extrapolations backward using the short-lived products as an estimate of production rate is an inadequate means of constructing the long time scale rates of nucleosynthesis. However it is certain that we are now looking through and resolving the granular structure of nucleosynthesis.

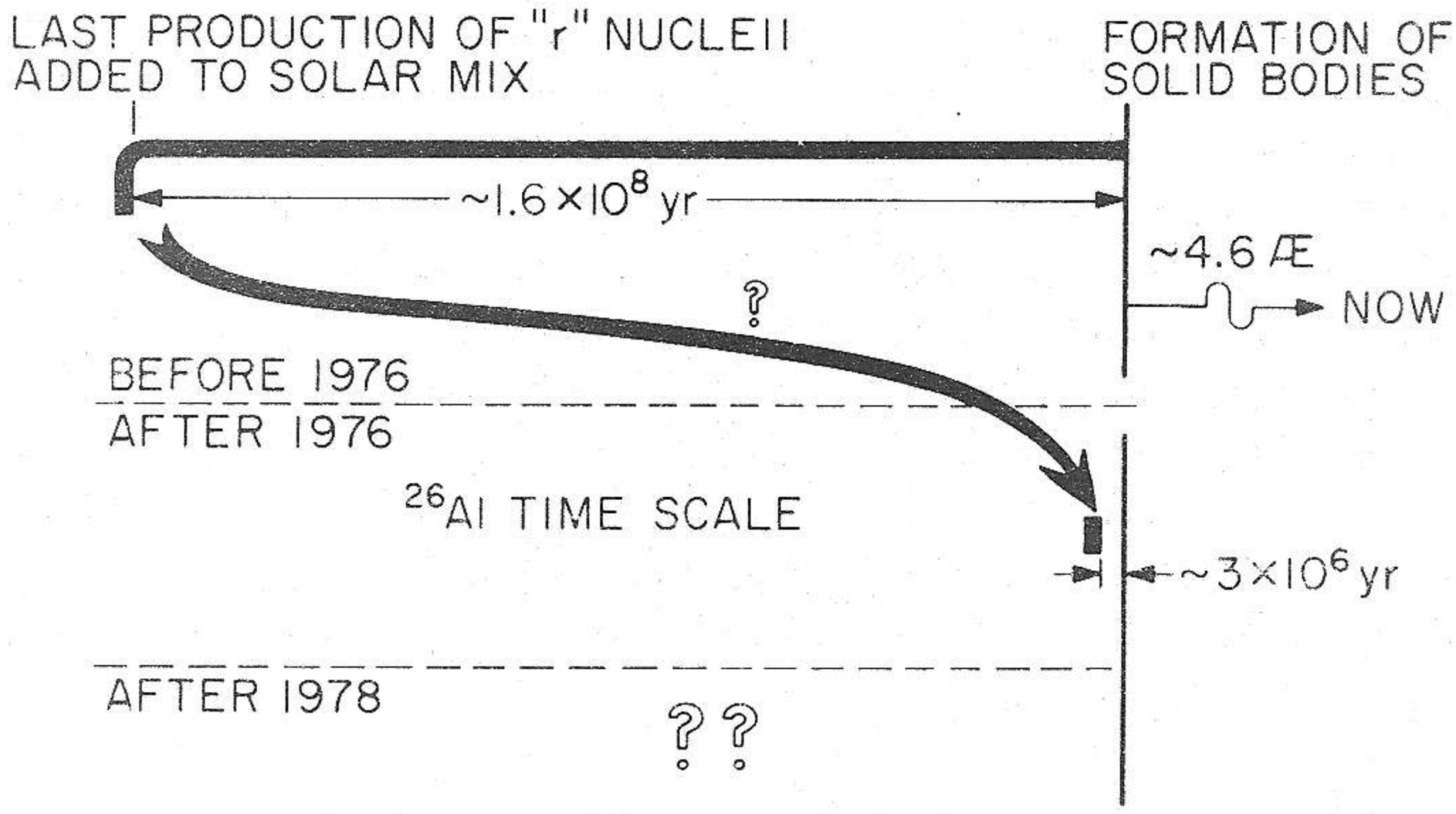

Fig. 23. Cartoon showing the required change in the length of the time interval between isolation of the solar system and condensation and aggregation of meteorites. Most plausible interpretation is the addition of ^{26}Al, ^{107}Pd, some ^{129}I, (^{135}Cs ?) within ~ 3×10^6 y of last production.

COMMON AND UNCOMMON ANOMALIES THE UNMADE CONNECTION

The FUN inclusions show isotopic anomalies in almost every refractory element so far measured. These FUN inclusions are relatively rare as they have been found to comprise only a few percent of the class of Ca-Al chondrules and aggregates. While the searches are not extensive, the isotopic anomalies found in the refractory elements in the two FUN inclusions are not observed in a variety of other meteorites. In contrast to this behavior, the isotopic anomalies observed for oxygen are common to almost all of the Ca-Al rich chondrules and aggregates. The relative fractional magnitudes of the oxygen effects are one to two orders of magnitude greater than found in the FUN samples for the refractory elements. Small but distinct differences in oxygen composition are found in a variety of meteorites. In general, samples which are found to have large oxygen effects are found to have no observable effects in other elements. It, therefore, appears that the isotopic anomalies in the FUN inclusions are not correlated with the widespread oxygen anomaly. The connection with the oxygen effects found in the FUN samples is seen in a departure from the typical mixing line between O_E and O_N and is most plausibly due to mass fractionation as is found for several elements. This observation is at present the only link between what otherwise appear to be distinctive phenomena. The other major element (in the solar system) that shows major and widespread isotopic anomalies is Ne. The Ne effects appear to be related

to differences in isotopic composition of the solar gas and that impregnated into dust (or solids). Because of the chemically inert nature of Ne clear correlations with condensible material are not apparent. Some of the observations in oxygen hint at differences between gas and dust. Certainly the degree of chemical reactivity between the different reservoirs or chemical compounds must be important in isolating them. From the magnitude of the oxygen effects (and possibly the Ne) it seems that the nuclear processes responsible for the O_E component must be extremely specific with only minor contributions to most other elements (both low and high *Z*). The exotic nuclides responsible for the UN effects in C-1 and EK-1-4-1 thus appear to come from a source distinct from that which either made or isolated O_E. At best it would appear that a connection between the common anomaly in oxygen and the uncommon anomalies in the refractory elements has not been made.

CONCLUSIONS

There is good reason to believe that continuing study of the late nucleosynthetic processes may give us better understanding of solar system formation, and direct insight in element processing-reprocessing in massive rapidly evolving stars.

The difficulty with any canonical astrophysical explanation (*e.g.*, a supernova) of the observations reviewed here is that no one has come forward with a plausible scenario that explains the observed isotopic anomalies or which clearly predicts new discoveries of isotopic anomalies. It is hoped that this report will provoke serious scholarly inquiry into the basic nuclear astrophysical processes that must be the cause of the isotopic effects. More intensive searches for samples with correlated isotope anomalies will be necessary. The demonstration of isotopic effects in a chain of elements which are directly related to nuclear reactions should prove a key to identifying the actual processes causing the anomalies. There are also major problems in our understanding of the condensation process and the chemical and ion reactions which occur between early and late formed parcels of matter in the early solar system. We will, no doubt, now begin to focus on the chemical « alteration » processes of « primitive » material — an issue which has been mostly ignored.

ADDENDUM

A variety of other intermediate to short-lived radioactive nuclear species may have been produced in the terminal event depending on the details of the particular nuclear processes involved. Searches for other extinct radioactivities are underway [*cf.*, for ^{41}Ca ($\tau_{1/2} = 1.3. \times 10^5$ y); BEGEMANN and STEGMANN, 1976]. Subsequent to the presentation at the Liège meeting, KELLY and WASSERBURG (1978) reported the discovery of 4 % excess of ^{107}Ag in an iron meteorite with a ratio $Pd/Ag \sim 10^4$. They consider this as most plausibly due to the decay of ^{107}Pd and infer $(^{107}Pd/^{110}Pd)_\odot \geq 2 \times 10^{-5}$. This is compatible with the ^{26}Al

time scale, the $(^{129}I/^{127}I)_{\odot}$ value, and the magnitude of the amount of addition ($\sim 10^{-4}$) of intermediate Z nuclei to the solar system. This implies also the formation of differentiated planetary bodies within $\sim 20 \times 10^6$ y of the last stage of nucleosynthesis. It is obvious that confirmation of this excess of ^{107}Ag and an adequate demonstration of the *in situ* decay of ^{107}Pd are urgently needed. The existence of ^{107}Pd directly connects the time of last nucleosynthesis and planetary differentiation while the ^{26}Al connects the last nucleosynthesis with formation of dust and marbles.

ACKNOWLEDGEMENTS

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The contributions of Malcolm McCulloch to part of the data summarized here merit special mention. Discussion with and stimulation from our colleagues in the Kellogg Radiation Laboratory is gratefully acknowledged. In this report we have omitted a general discussion of the rare gases and, in particular, xenon and krypton anomalies. These observations and ideas are of great importance and have been the subject of extensive investigation and discussion over the past two decades. It is hoped that our noble colleagues with affection for atomic numbers 36-10-54 will recognize the limited nature of this report.

REFERENCES

ALEXANDER, E.C. Jr., LEWIS, R.S., REYNOLDS, J.H. and MICHEL, M.C., 1971, ^{244}Pu: confirmation of an extinct radioactivity, *Science,* **172**, 837-840.

ALLEN, B.J., GIBBONS, H.H. and MACKLIN, R.L., 1971, Nucleosynthesis and neutron capture cross sections. *Adv. Nucl. Phys.*, **4**, 205-259.

ARNETT, W.D. and WEFEL, J.P., 1978, ^{26}Al production from a stellar evolutionary sequence, *Ap. J. (Letters),* **224**, 139-142.

BEGEMANN, F., GEISS, J. and HESS, D.C., 1957, Radiation age of a meteorite from cosmic ray produced ^{3}He and ^{3}H. *Phys. Rev.*, **107**, 540-542.

BEGEMANN, F. and STEGMANN, W., 1976, Implications from the absence of a ^{41}K anomaly in an Allende inclusion, *Nature,* **259**, 549-550.

BLACK, D.C., 1972, On the origins of trapped He, Ne, and Ar isotopic variations in meteorites - II. Carbonaceous meteorites, *Geochim. Cosmochim. Acta,* **36**, 377-394.

BLANDER, M. and FUCHS, L.H., 1975, Ca-Al-rich inclusions in the Allende meteorite: evidence for a liquid origin. *Geochim. Cosmochim. Acta,* **39**, 1605-1619.

BRADLEY, J.G., HUNEKE, J.C., and WASSERBURG, G.J. 1978, Ion microprobe evidence for the presence of excess ^{26}Mg in an Allende anorthite crystal. *J. Geophys. Res.*, **83**, 244-254.

BURBIDGE, E.M., BURBIDGE, G.R., FOWLER, W.A. and HOYLE, F., 1957, Synthesis of the elements in stars. *Rev. Mod. Phys.*, **29**, 547-650.

CAMERON A.G.W., 1957, Nuclear reactions in stars and nucleogenesis. *Pub. A.S.P.*, **69**, 201-222.

CAMERON, A.G.W, 1963, Formation of the solar nebula, *Icarus,* **1**, 339-342.
——, 1973a, Abundances of the elements in the solar system, *Space Sci. Rev.*, **15**, 121-146.
——, 1973b, Are large time differences in meteorite formation real?, *Nature,* **246**, 30-32.
CHRISTOPHE-MICHEL-LEVY, M., 1968, Un chondre exceptionnel dans la météorite de Vigarano. *Bull. Soc. Fr. Minéral. Cristallogr.*, **91**, 212-214.
CLAYTON, D.D., 1975, Extinct radioactivities: trapped residuals of presolar grains, *Ap. J.*, **199**, 765-769.
CLAYTON, R.N., GROSSMAN, L. and MAYEDA, T.K., 1973, A component of primitive nuclear composition in carbonaceous meteorites, *Science,* **182**, 485-488.
CLAYTON, R.N. and MAYEDA, T.K., 1977, Correlated O and Mg isotope anomalies in Allende inclusions I. Oxygen, *Geophys. Res. Lett.*, **4**, 295-298.
——, 1978, Genetic relations between iron and stony meteorites. *Earth Planet. Sci. Letters,* **40**, 168-174.
CLAYTON, R.N., MAYEDA T.K. and EPSTEIN, S., 1978, Isotopic fractionation of Si in Allende inclusions, In *Proc. 9th Lunar Science Conf.*, Houston, pp. 1267-1278.
CLAYTON, R.N., ONUMA, N., GROSSMAN, L. and MAYEDA, T.K., 1977, Distribution of the pre-solar component in Allende and other carbonaceous chondrites, *Earth Planet. Sci. Lett.*, **34**, 209-224.
CLAYTON, R.N., ONUMA, N. and MAYEDA, T.K., 1976, A classification of meteorites based on oxygen isotopes. *Earth Planet. Sci. Lett.*, **30**, 10-28.
CONRAD, J., 1976, Analysis of the *s*-process and the nuclear synthesis of the elements, Ph. D. Thesis, Heidelberg.
CONSOLMAGNO, G.J., 1977, The magnetic Reynolds number of the solar nebula, *Meteoritics,* **12**, 200.
DEARBORN, D., TINSLEY, B.M. and SCHRAMM D.N., 1978, On the origin and evolution of isotopes of carbon, nitrogen and oxygen., *Ap. J.*, **223**, 557-566.
EBERHARDT, P., 1974, A neon-E rich phase in the Orgueil carbonaceous chondrite, *Earth Planet. Sci, Lett.*, **24**, 182-187.
——, 1978, A Ne-E rich phase in Orgueil: results of stepwise heating experiments. In *Proc. 9th Lunar Sci. Conf.*, Houston, 1027-1051.
ESAT, T.M., LEE, T., PAPANASTASSIOU, D.A. and WASSERBURG, G.J., 1978, Search for ^{26}Al effects in the Allende FUN inclusion Cl. *Geophys. Res. Lett.*, **5**, 807-810.
FOWLER, W.A., GREENSTEIN, J.L. and HOYLE, F., 1962, Nucleosynthesis during the early history of the solar system. *Geophys. J.*, **6**, 148-220.
GEISS, J., BUEHLER, F., CERUTTI, H., EBERHARD, P. and FILLEUX, CH., 1972, Solar wind composition experiment, *Apollo 16 Preliminary Science Report,* NASA SP-315, section 14-1.
GOLDREICH, P. and WARD, W.R., 1973, Formation of planetesimals. *Ap. J.*, **183**, 1051-1061.
GRAY, C.M. and COMPSTON, W., 1974, Excess ^{26}Mg in the Allende meteorite, *Nature,* **251**, 495-497.
GRAY, C.M., PAPANASTASSIOU, D.A. and WASSERBURG, G.J., 1973, The identification of early condensates from the solar nebula, *Icarus,* **20**, 213-239.
GROSSMAN, L., 1972, Condensation in the primitive solar nebula. *Geochim. Cosmochim. Acta,* **36**, 597-619.
——, 1975, Petrography and mineral chemistry of Ca-rich inclusions in the Allende meteorite. *Geochim. Cosmochim. Acta,* **39**, 433-454.

HOHENBERG, C.M., 1970, Xe from the Angra dos Reis meteorite, *Geochim. Cosmochim. Acta,* 34, 185-191.

HOLMES, J.A., WOOSLEY, S.E., FOWLER, W.A. and ZIMMERMAN, B.A., 1976, Tables of thermonuclear reaction rate data for neutron induced, reactions of heavy nuclei. *At. Data Nucl. Data Tables,* 18, 305.

HOWARD, W.M., ARNETT, W.D., CLAYTON, D.D. and WOOSLEY, S.E., 1972, Nucleosynthesis of rare nuclei from seed nuclei in explosive carbon burning. *Ap. J.*, 175, 201-216.

HUTCHEON, I.D., STEELE, I.M., SMITH, J.V. and CLAYTON, R.N., 1978, Ion microprobe, electron microprobe and cathodoluminescence data for Allende inclusions with emphasis on plagioclase chemistry, *Proc. 9th Lunar Planet. Sci. Conf.*, pp. 1345-1368.

JEFFERY, P.M. and REYNOLDS, J.H., 1961, Origin of excess ^{129}Xe in stone meteorites, *J. Geophys. Res.*, 66, 3582-3583.

KELLY, W.R. and WASSERBURG, G.J., 1978, Evidence for the existence of ^{107}Pd in the early solar system. *Geophys. Res. Lett.*, 5, 1079-1082.

LEE, T., 1978, A local proton irradiation model for isotopic anomalies in the solar system. *Ap. J.*, 224, 217-226.

LEE, T. and PAPANASTASSIOU, D.A., 1974, Mg isotopic anomalies in the Allende meteorite and correlation with O and Sr effects. *Geophys. Res. Lett.*, 1, 225-228.

LEE, T., PAPANASTASSIOU, D.A. and WASSERBURG, G.J., 1976, Demonstration of ^{26}Mg excess in Allende and evidence for ^{26}Al. *Geophys. Res. Lett.*, 3, 109-112.

——, 1977, ^{26}Al in the early solar system: fossil or fuel? *Ap. J. (Letters),* 211, 107-110.

——, 1978, Ca isotopic anomalies in the Allende meteorite, *Ap. J. (Letters),* 220, 21-25.

LEE, T., RUSSELL, W.A. and WASSERBURG, G.J., 1979, Ca isotopic anomalies and the lack of ^{26}Al in an unusual Allende inclusion. *Ap. J., (Letters)* in press.

LEWIS, R.S., 1975, Rare gases in separated whitlockite from the St. Severin chondrite: Xe and Kr from fission of extinct ^{244}Pu, *Geochim. Cosmochim. Acta,* 39, 433-454.

LORD, H.C. III, 1965, Molecular equilibria and condensation in a solar nebula and cool stellar atmospheres. *Icarus,* 4, 279-288.

LORIN, J.-C. and CHRISTOPHE MICHEL-LEVY, M., 1978, Radiogenic ^{26}Mg fine scale distribution in Ca-Al inclusions of the Allende and Leoville meteorites. In *Short Papers of the 4th ICGCIG,* Denver, 257-259.

LORIN, J.-C., SHIMIZU, N., CHRISTOPHE MICHEL-LEVY M. and ALLEGRE, C.J., 1977, The Mg isotope anomaly in carbonaceous chondrites; an ion probe study, *Meteoritics,* 12, 299-300.

LUGMAIR, G.W., MARTI, K. and SCHEININ, N.B., 1978, Incomplete mixing of products from *r*, *p*, and *s*-process nucleosynthesis: Sm-Nd systematics in Allende inclusion EK-1-04-1. In *Lunar and Planetary Science IX,* Lunar and Planetary Institute, Houston, pp. 672-674.

MARVIN, U.B., WOOD, J.A. and DICKEY, J.S. Jr., 1970, Ca-Al rich phases in the Allende meteorite, *Earth Planet. Sci. Lett.*, 7, 346-350.

McCULLOCH, M.T. and WASSERBURG, G.J., 1978a, Ba and Nd isotopic anomalies in the Allende meteorite. *Ap. J. (Letters),* 220, 15-19.

——, 1978b, More anomalies from the Allende meteorite: Samarium. *Geophys. Res. Lett.*, 5, 599-602.

MUSGROVE, A.R. de L., BODLMAN, J.W. and MACKLIN, R.L., 1976, KeV neutron capture cross sections of ^{134}Ba and ^{136}Ba. *Nucl. Phys.*, **A256**, 173-188.

NIEMEYER, S., 1978, I-Xe dating of inclusions from IAB iron meteorites. In *Lunar and Planetary Science IX,* Houston, 808-810.

NIER, A.O., McELROY, M.B. and YUNG, Y.L., 1976, Isotopic composition of the Martian atmosphere. *Science,* **194**, 68-70.

PAPANASTASSIOU, D.A., HUNEKE, J.C., ESAT, T.M. and WASSERBURG, G.J., 1978, Pandora's Box of the nuclides. In *Lunar and Planetary Science IX,* Lunar and Planetary Institute, Houston, pp. 859-861.

PAPANASTASSIOU, D.A., LEE, T. and WASSERBURG, G.J., 1977, Evidence for ^{26}Al in the solar system. In *Comets, Asteroids, Meteorites,* ed. A.H., DELSEMME (Univ. of Toledo), 343-349.

PAPANASTASSIOU, D.A. and WASSERBURG, G.J., 1978, Strontium isotopic anomalies in the Allende meteorite. *Geophys. Res. Lett.*, **5**, 595-598.

PODOSEK, F.A, 1970, The abundance of ^{244}Pu in the early solar system. *Earth and Planet. Sci. Lett.*, **8**, 183-187.

——, 1972, Cas retention chronology of Petersburg and other meteorites. *Geochim. Cosmochim. Acta,* **36**, 755-772.

REYNOLDS, J.H., 1960, Determination of the age of the elements. *Phys. Rev. Lett.*, **4**, 8-10.

——, 1963, Xenology. *J. Geophys. Res.*, **68**, 2939-2956.

ROWE, M.W. and KURODA, P.K., 1965, Fissiogenic Xe from the Pasamonte meteorite. *J. Geophys. Res.*, **70**, 709-714.

RUSSELL, W.A., PAPANASTASSIOU, D.A. and TOMBRELLO, T.A., 1978, Ca isotope fractionation on the Earth and other solar system materials. *Geochim. Cosmochim. Acta,* **42**, 1075-1090.

SAFRONOV, V.S., 1969, Evolution of the protoplanetary cloud and the formation of the earth and planets (in Russian). Nauka Moscow. Engl. transl. as NASA TT F-677. NTIS, Springfield, VA (1972).

SCHRAMM, D.N. and WASSERBURG, G.J., 1970, Nucleochronologies and the mean age of the elements. *Ap. J.*, **162**, 57-69.

TAYLOR, H.P., DUKE, M.B., SILVER, L.T. and EPSTEIN, S., 1965, Oxygen isotope studies of minerals in stony meteorites. *Geochim. Cosmochim. Acta,* **29**, 489-512.

TERA, F., PAPANASTASSIOU, D.A. and WASSERBURG, G.J. 1974, Isotopic evidence for a terminal lunar cataclysm. *Earth Planet. Sci. Lett.*, **22**, 1-21.

TRURAN, J.W. and CAMERON, A.G.W., 1978, ^{26}Al production in explosive carbon burning. *Ap. J.*, **219**, 226-229.

TRURAN, J.W., COWAN, J.J. and CAMERON, A.G.W., 1978. The He-driven *r*-process in supernovae. *Ap. J. (Letters),* **222**, 63-67.

VOSHAGE, H., 1978, Investigations on cosmic ray produced nuclides in iron meteorites, 2. New results on ^{41}K/^{40}K - ^{4}He/^{21}Ne exposure ages and the interpretation of age distributions. *Earth Planet. Sci. Lett.*, **40**, 83-90.

WANNIER, P.G., LUCAS, R., LINKE, R.A., ENCRENAZ, P.J., PENZIAS, A.A. and WILSON, R.W., 1976, The abundance ratio ^{17}O/^{18}O in dense interstellar clouds, *Ap. J. Lett.*, **205**, 169-171.

WARD, R.A., NEWMAN, M.J. and CLAYTON, D.D., 1976, *s*-process studies: branching and the time scale. *Astrophys. J. Suppl.*, **31**, 33-59.

WARK, D.A. and LOVERING, J.F., 1977, Marker events in the early evolution of the solar system: evidence from rims on Ca-Al-rich inclusions in carbonaceous chondrites. *Proc. 8th Lunar Science Conf.*, Houston, pp. 95-112.

WASSERBURG, G.J., 1978, Proceedings R.A. Welch Foundation Conf. on Chem. Res. XXI Cosmochemistry, Nov. 7-9, 1977, ed. W.O. Milligan, Houston, pp. 95-100.

WASSERBURG, G.J., HUNEKE, J.C. and BURNETT, D.S., 1969, Correlation between fission tracks and fission type Xe in meteoritic whitlockite, *J. Geophys. Res.*, **74**, 4221-4232.

WASSERBURG, G.J., LEE, T. and PAPANASTASSIOU, D.A., 1976, Large ^{26}Mg excesses in an Allende inclusion and a ^{26}Al-^{26}Mg internal isochron. *Meteoritics,* **11**, 384.

——, 1977, Correlated O and Mg isotopic anomalies in Allende inclusions: II Magnesium. *Geophys. Res. Lett.*, **4**, 299-302.

WASSERBURG, G.J., TERA, F., PAPANASTASSIOU, D.A. and HUNEKE, J.C., 1977, Isotopic and chemical investigations on Angra dos Reis. *Earth Planet. Sci. Lett.*, **35**, 294-316.

UREY, H.C., 1955, The cosmic abundances of potassium, uranium, and the heat balance of the Earth, the Moon, and Mars, *Proc. Nat. Acad. Sci. U.S.*, **41**, 127-144.

WOOSLEY, S.E., ARNETT, W.D. and CLAYTON, D.D., 1973, The explosive burning of oxygen and silicon. *Ap. J. Suppl.*, **26**, 231-312.

WOOSLEY, S.E. and HOWARD, W.M., 1978, The *p*-process in supernovae. *Ap. J. Suppl. Series,* **36**, 285-304.

YEH, H.-W. and EPSTEIN, S., 1978, $^{29}Si/^{28}Si$ and $^{30}Si/^{28}Si$ of meteorites and Allende inclusions. In *Lunar and Planetary Science IX,* Lunar and Planetary Institute, Houston, 1289-1291.